

Space, Plasma and Climate Community
Department of Physics
Imperial College London

Exploiting far-infrared signals of water vapour and ice clouds

**A new retrieval capability developed in support of the ESA
FORUM mission**

Sanjeevani Panditharatne

Supervised by Prof. Helen Brindley and Dr. Caroline Cox

Submitted in part fulfilment of the requirements for the PhD
degree at Imperial College London

April 2025

Abstract

This thesis presents the extension of the Rutherford Appleton Laboratory Infrared Microwave Sounding (IMS) optimal estimation retrieval scheme into the far-infrared in preparation for the European Space Agency's Far-infrared Outgoing Radiation Understanding and Monitoring (FORUM) mission. Studies have indicated that upwelling far-infrared radiances ($100\text{--}667\text{ cm}^{-1}$) hold significant information about ice cloud microphysics, and the distribution of water vapour. This work has extended IMS to exploit these far-infrared signals, and applied it to observations of upwelling far-infrared radiances that have been adapted to mimic the FORUM Sounding Instrument's (FSI) spectral characteristics. The work in this thesis is presented in three stages.

First, the retrieval scheme has been optimised for clear-sky retrievals from simulated FSI radiances, and compared against the existing IMS configuration for the Infrared Atmospheric Sounding Interferometer (IASI). Comparable retrieval biases were observed for temperature and water vapour, however there was an increase of ~ 1 degree of freedom for water vapour and temperature for the FSI configuration.

Second, IMS has been applied to eight clear-sky observations of upwelling far- and mid-infrared radiances taken during the PIKNMIX-F campaign. Retrievals from these radiances using the modified code showed a strong agreement with contemporaneous in-situ measurements of the atmospheric state, reducing the RMSE by 18% for water vapour from the a-priori, giving confidence in its performance.

Finally, IMS has been used to perform the first retrieval of ice cloud properties (cloud optical thickness, cloud effective radius, cloud top height, ice crystal habit) from an airborne observation of upwelling far- and mid-infrared radiances taken above a cirrus cloud layer. Despite the known limitations of IMS, the retrieved values agree with in-situ measurements of the cloud. The inclusion of the far-infrared enables a distinction between two habits that was not possible using only the mid-infrared ($667\text{--}2000\text{ cm}^{-1}$), and halves the uncertainties in the retrieved outputs.

Acknowledgements

My first thanks are to my supervisors, Helen Brindley and Caroline Cox, for both their guidance and the opportunities they have provided me with. Without them, this project would have been impossible. Special thanks goes to Laura Warwick and Jon Murray, who have remained scientific and instrument gurus for me throughout my PhD, even at 2 am in the icy mountains of Norway. I would also like to thank Richard Siddans, who allowed me to work on the Infrared Microwave Sounding retrieval scheme; Rich Bantges, who was always willing to sound out my ice crystal ideas thoughts; and Lorenzo Matteini and Paulo Ceppi, who provided feedback throughout my time at Imperial.

Next, I would like to thank all the teams who worked on TAFTS and ARIES, as well as Rui Song, who promptly analysed all of the lidar data for me, and the FAAM team who were kind enough to accommodate me on a test flight. This thanks extends to those at the Grantham Institute who were always so supportive.

I am extremely grateful to my friends in SPLAT and NCEO, who have kept me sane over the last few years, as well as Natalie and Ostin, who I hope enjoyed my lengthy voice notes.

Finally, I dedicate this thesis to the love of my life Sam, who is the sunshine in my life, and my mother, Wasana, who believed in me from the start.

Statement of Originality

The following thesis is the product of my own research. All results or information taken from other work has been acknowledged and referenced appropriately.

Copyright Declaration

The copyright of this thesis rests with the author. Unless otherwise indicated, its contents are licensed under a Creative Commons Attribution-Non Commercial 4.0 International Licence (CC BY-NC). Under this licence, you may copy and redistribute the material in any medium or format. You may also create and distribute modified versions of the work. This is on the condition that: you credit the author and do not use it, or any derivative works, for a commercial purpose. When reusing or sharing this work, ensure you make the licence terms clear to others by naming the licence and linking to the licence text. Where a work has been adapted, you should indicate that the work has been changed and describe those changes. Please seek permission from the copyright holder for uses of this work that are not included in this licence or permitted under UK Copyright Law.

Contents

Abstract	2
List of Tables	8
List of Figures	10
List of Abbreviations	20
1 Introduction	22
1.1 Earth's Radiation Budget	22
1.1.1 The Role of Water Vapour	25
1.1.2 The Role of Cirrus Clouds	26
1.2 The Far-Infrared	27
1.2.1 Instruments In The Far-Infrared	28
1.2.2 The Far-Infrared Outgoing Radiation Understanding and Monitoring Mission	31
1.3 Exploiting the Far-Infrared in Retrievals	32
1.4 Structure of Thesis	34
2 The Retrieval Method	37
2.1 Radiative Transfer	37
2.1.1 Atmospheric Absorption and Emission	40
2.1.2 The Scattering Effects of Ice Clouds	41
2.1.3 Radiative Transfer Models	47
2.2 Optimal Estimation Retrievals	49
2.2.1 Characterising the retrievals	55
2.2.2 A comparison of the retrieved state to the a-priori state	55
2.2.3 A comparison of the retrieved state to the true state	57
2.2.4 Additional Retrieval Diagnostic Parameters	59
3 Building FORUM-aircraft Observations	61
3.1 Fourier Transform Spectroscopy	61
3.2 Instruments Used in This Work	65
3.3 Constructing FORUM-aircraft Observations	70
4 Extending the Infrared Microwave Sounding Retrieval Scheme into the Far-Infrared	77
4.1 Introduction	78
4.2 Infrared Microwave Sounding (IMS)	78
4.2.1 The State Vector	80

4.2.2	Radiative Transfer for TOVS (RTTOVv12)	82
4.2.3	Default Channel Selection	83
4.3	Development Outline	84
4.4	Simulated Clear-Sky Test Cases	85
4.5	Evaluating the Current Ability of IMS	87
4.6	Technical Extension for the FORUM Sounding Instrument	89
4.6.1	Using the FSI Clear-Sky Regression Coefficients	90
4.6.2	Building the New Covariance Matrices	91
4.7	Optimising for Clear-Sky Retrievals from FSI Radiances	97
4.8	Conclusions	102
5	Application to Clear-Sky Observations from the Upper Troposphere	105
5.1	Introduction	106
5.2	Flight C153 Overview	106
5.2.1	Auxiliary Information	107
5.3	Selection and Preparation for the Radiance Observations	109
5.4	Channel Selection and Testing on Simulations	113
5.5	Retrievals from Aircraft Observations	116
5.5.1	Initial Retrieval	116
5.5.2	Revised Retrievals	119
5.6	Conclusions	126
6	First Retrievals of Ice Cloud Properties from Far-Infrared Observations	129
6.1	Introduction	130
6.2	Approximations Used to Retrieve Ice Cloud Properties	130
6.2.1	Simulating the Radiances	131
6.2.2	Retrieval Approximations	134
6.3	B895 Flight Overview	135
6.3.1	Cloud Instrumentation	138
6.3.2	Estimating the CER	140
6.4	Optimising IMS for Ice Cloud Retrievals from the B895 Flight	143
6.4.1	Constructing Other Ice Crystal Habit Coefficients	143
6.4.2	Retrieving the Ice Crystal Habit	148
6.4.3	Optimal Estimation Retrieval Constraints	149
6.4.4	Channel Selection	150
6.5	Fixed Temperature and Water Vapour Retrievals	152
6.5.1	Retrieval from the simulation	153
6.5.2	Initial retrieval from the observation	155
6.6	Retrieval of Temperature, Water Vapour, and Cloud Properties	157
6.6.1	Retrieval of the Ice Crystal Habit	157
6.6.2	Retrieval of Temperature and Water Vapour	159
6.6.3	Retrieval of COT, CER, and CTH	161
6.7	Conclusions	163
7	Conclusions and Future Work	165
7.1	Suggestions for Future Research	170
	Appendix A: Glossaries of key symbols	173

Appendix B: Water vapour conversions	175
Bibliography	177

List of Tables

1.1	A summary of the spectral ranges and resolution for each instrument as well as their measurement of the upwelling or downwelling radiation (Mlynckzak et al. 2006, Bianchini et al. 2019, Canas et al. 1997, Palchetti et al. 2021, Murray et al. 2024, Hanel et al. 1970, 1971, Kempe et al. 1980, L'Ecuyer et al. 2021). The spectral resolution for PREFIRE is specified in wavelength.	29
4.1	A summary of the retrieval targets within the state vector in IMS. For the COT, the a-priori is stored in log form and the covariance represents the variation on the log of the COT.	81
4.2	A summary of the median and median absolute deviation in diagnostic parameters for the retrievals from the 240 test cases using the IASI configuration. The absolute residual skin temperature and surface emissivity residual were calculated by finding the median of the absolute difference between the retrieved and 'true' values. The remainder of the diagnostic parameters are described in Section 2.2 with definitions in Table A2.	89
4.3	A summary of the median and median absolute deviation in diagnostic parameters for retrievals from the 240 test cases using the IASI and FSI configurations.	99
5.1	The times of each dropsonde release and the closest TAFTS scan in time from the C153 flight on 13 March 2019. For each TAFTS scan, the closest 6 ARIES scans have been averaged to provide the best match for the field of view and scan time between the instruments. The time of the central ARIES scan is shown.	110
5.2	The RMSE value relative to each AK-treated dropsonde profile for temperature and water vapour for the 8 observations against the a-priori (ERA5) and retrieved profiles. To aid later interpretation, RMSE values have been separated into above and below 5 km, alongside those for the full profile.	122
6.1	The timings of relevant measurements taken during the B895 flight.	136
6.2	The relative standard deviation (%) in each parameter fitted in the bimodal Gaussian: $f(x) = A_1 \exp\left[\frac{-(x-\mu_1)^2}{2\sigma_1^2}\right] + A_2 \exp\left[\frac{-(x-\mu_2)^2}{2\sigma_2^2}\right]$ corresponding to the red dashed curves in Figure 6.5.	141
6.3	The CER calculated from the PSDs measured by the Cloud 2-DS probe, and the habit distributions measured by the 3V-CPI for temperature regions shown in Figure 6.4. The temperatures have been converted to altitudes using the Dropsonde 1 measurements (Figure 6.4c) and have been used to calculate the approximate times and distances since and from the radiance observation.	142
6.4	The 4 different retrieval configurations used: with and without the far-infrared, and assuming the SC or GHM bulk optical property model.	149

6.5	The retrieved COT, CER, and CTH from the LBLDIS GHM FORUM-aircraft simulation described in Section 6.5.1 in comparison to the input values for the simulation. Uncertainties shown are one ESD. J_y is also shown for each retrieval configuration and is the same as in Figure 6.12a.	155
A1	Glossary of key symbols described Section 2.1.	173
A2	Glossary of key symbols described Section 2.2.	174

List of Figures

1.1	The radiative fluxes associated with the Earth's globally averaged radiation budget from 2006-2018. Numbers indicate the best estimates for the magnitudes in Wm^{-2} , with uncertainties shown in parentheses (5-95 % confidence range). Taken from Forster et al. (2021)	23
1.2	Example spectra simulated using a radiative transfer code for a tropical profile in a clear sky (orange) and with an example ice cloud in the upper troposphere (blue) atmosphere. The blackbody curve for the surface temperature is shown in black.	28
1.3	The satellite ground tracks and footprints for the FSI (red), FEI (yellow), and IASI-NG (blue). Taken from Palchetti et al. (2020)	32
2.1	The transmittance and absorption of six strongly absorbing gases in the infrared, as well as for the six gases combined in the absence of clouds. Taken from Andrews (2010).	40
2.2	Ice crystal habits in ice clouds: (a) pristine hexagonal column, (b) pristine hexagonal plate, (c) six-branched bullet-rosette, (d) six-branched bullet-rosette with conical air cavities within each branch, (e) polycrystal, (f) hexagonal ice aggregate, (g) Inhomogeneous Hexagonal Monocrystal, (h) hexagonal chain aggregate, (i) rosette-aggregate, (j) droxtal, (k) Gaussian random sphere and (l) Chebyshev polynomial. Taken from Baran (2009).	42
2.3	The link between state and measurement space is used to solve the general inverse problem. The forward model translates the measurement vector (y) into state space. The associated measurement error is propagated into a region of possible solutions. The true state x should lie within this region, and the estimated $\hat{x}$ must capture it within its uncertainties. Taken from Rodgers (2000).	50
3.1	A schematic of a two-beam Michelson interferometer.	62
3.2	A summary of the uncertainties related to radiance observations taken by TAFTS and ARIES that are used in Chapter 5. (a) An example of a clear-sky spectrum (single scan) observed by TAFTS and ARIES during the C153 flight. (b) The Noise-Equivalent-Spectral-Radiance (NESR) and (c) Absolute Radiometric Accuracy (ARA) for TAFTS and ARIES from Warwick et al. (2022), as well as the expected NESR values for the FSI. The target ARA for the FSI described is in brightness temperature, and is 0.1 K between 300-1100 cm^{-1} , 0.2 K between 200-300 cm^{-1} and 1100-1300 cm^{-1} , and 1 K elsewhere.	68

3.3	A summary of the uncertainties related to radiance observations taken by TAFTS and ARIES that are used in Chapter 6. (a) An example of a cloudy spectrum (single scan) observed by TAFTS and ARIES during the B895 flight. (b) The Noise-Equivalent-Spectral-Radiance (NESR) and (c) Absolute Radiometric Accuracy (ARA) for TAFTS and ARIES from Bantges et al. (2020), as well as the expected NESR values for the FSI. The target ARA for the FSI described is in brightness temperature, and is 0.1 K between 300-1100 cm^{-1} , 0.2 K between 200-300 cm^{-1} and 1100-1300 cm^{-1} , and 1 K elsewhere.	69
3.4	Instrument line shapes for (green) ARIES, (blue) TAFTS, and (red) FSI that are used in this work. The instrument line shapes are offset for TAFTS (+2) and the FSI (+1) for easier comparison.	71
3.5	The process used to develop FORUM-aircraft observations or simulations. The whole flowchart is used to calculate the error associated with the apodisation process. The path on the right-hand side describes the 'direct' process of applying the FSI apodisation to a high-resolution simulated spectrum, where radiances aren't modified to look like TAFTS and ARIES first. The path on the left-hand side describes the 'indirect' process where the TAFTS and ARIES spectral characteristics are applied, the TAFTS spectral characteristics are removed, and then the FSI apodisation is applied. The steps applied to the TAFTS and ARIES observations are outlined in purple.	72
3.6	The uncertainty associated with developing FORUM-aircraft observations in radiance units. The light blue line (clear-sky) and purple line (cloudy) show the residuals between LBLRTMv12.11 or LBLDIS simulations of the TAFTS and ARIES observations that have had the FSI apodisation directly applied and been made to look like (a) TAFTS or (b) ARIES first. These simulations are based on observations from Warwick et al. (2022) and Bantges et al. (2020). The green line shows the clear-sky residual where the simulated ARIES spectrum has a finer sampling (Fine Samp.) than in the real case before the FSI apodisation has been applied. The black line is the noise and calibration uncertainty combined in quadrature associated with the ARIES longwave channel and both TAFTS channels from Figure 3.3b and c for the B895 flight.	74
4.1	A schematic summarising the optimal estimation process within IMS. The observed spectrum and a-priori to the state are input into IMS. Using the radiative transfer model (RTTOVv12) the retrieval framework simulates a first guess spectrum. Components of RTTOVv12 are highlighted in green and described in Section 4.2.2. This spectrum is compared against the measured spectra at certain wavenumbers (channels), and a cost function is calculated. IMS works to minimise this cost function by adjusting the state to ultimately output the retrieved state and fitted spectrum. Components highlighted in red are spectrally dependent.	79

4.2	An example RTTOVv12 simulation of a spectrum measured by the IASI instrument overlaid with the 181 channels used within IMS to perform retrievals. The channels selected by ECMWF for use in Numerical Weather Prediction models are shown in green, and the additional channels selected for improving ozone and methane retrievals in IMS are shown in blue. The square-root-diagonal of S_y for IASI (Uncertainty) is also shown in red and is derived from residuals between IASI observations and RTTOVv12 simulations as described in Section 4.2.	83
4.3	(a) The spatial distribution of the 240 cases selected for testing IMS. Profiles were derived from MERRA-2. Tropical Cases: 70. Mid-Latitude Cases: 95. Arctic Cases: 75. The percentage of total cases with each (b) skin temperature, and (c) total column water vapour (TCWV) in the MERRA-2 reanalysis dataset for one day in March, June, September, and December in 2019 and used in the test dataset, shown in blue and red, respectively. Overlapping regions are shown in purple where the same proportion of cases is present in the test set and MERRA-2 reanalysis dataset.	86
4.4	The median (a-d) water vapour (WV) and (e-h) temperature (T) bias of the retrieved profile to the ‘true’ state across the testing cases for the optimised IASI configuration in red, in comparison to the a-priori bias to the ‘true’ state in orange. Biases are evaluated between 300 and 1000 hPa in 100 hPa bins as in Trent et al. (2023) and shown for (Global) all 240 cases as well as in separate latitudinal regions of (Polar) 60 to 90 degrees N/S, (Mid-Latitudes) 30 to 60 degrees N/S, and (Tropical) 30 N to 30 S. The water vapour percentage bias is calculated from absolute values in ppmv. Dashed lines show the median estimated standard deviation (ESD) for a single retrieval for the test cases in each bin. For water vapour this is plotted with an offset of -10 % for clarity.	88
4.5	The median (a) water vapour (WV) and (b) temperature (T) bias of the retrieved profile to the ‘true’ state across the testing cases for three different retrieval configurations. The retrievals using the IASI configuration and 181 channels is shown in red and is the same as in Figure 4.4. The retrievals using the IASI configuration with 178 channels is shown in green. The retrievals using the FSI configuration with the FSI channels closest to the 178 IASI channels is shown in light blue. The water vapour percentage bias is calculated from absolute values in ppmv. Dashed lines show the median estimated standard deviation (ESD) for a single retrieval for the test cases in each bin. For water vapour this is plotted with an offset of - 10% for clarity.	91
4.6	(a) The main diagonal of the IASI measurement covariance in red and the main diagonal of the FSI measurement covariance separated into components from the noise-equivalent-spectral-radiance (NESR) in blue and the target absolute radiometric accuracy (ARA) in green. As the ARA is provided in brightness temperature, it has been converted into radiance units for each of the ECMWF 83 profiles. The median is shown here, with the shading representing the minimum and maximum values. The square-root of the measurement covariances for the (b) NESR, (c) ARA, and (d) final combined matrix are also shown.	93

4.7	The median (a) water vapour (WV) and (b) temperature (T) bias of the retrieved profile relative to the ‘true’ state across the testing cases for three different retrieval configurations. The retrievals using the FSI configuration with the FSI channels closest to the 178 IASI channels is shown in light blue (FSI 178). The retrievals using the FSI configuration with the FSI channels and measurement covariance closest to the 178 IASI channels is shown in blue (FSI 178 S_y). The water vapour percentage bias is calculated from absolute values in ppmv. Dashed lines show the median estimated standard deviation (ESD) for a single retrieval for the test cases in each bin. For water vapour this is plotted with an offset of - 10% for clarity.	94
4.8	The different far-infrared surface emissivities contained within the Huang et al. (2016) dataset prescribed for different surface types. The Desert (Mean) surface emissivity corresponds to the average of the different desert surface types contained within the model that have different silt sizes. The surface emissivity for grass is partially obscured by the surface emissivity for conifers. Uncertainties for these emissivities are not provided with this database and the accuracy of each surface emissivity in the far-infrared has not been thoroughly validated.	96
4.9	The a-priori covariance for the surface emissivity for the FSI configuration. Above 645 cm^{-1} , the original IMS covariance has been interpolated onto the FSI spectral grid. Below 645 cm^{-1} , the covariance has been derived from a combination of the (sea) UWIREMIS atlas and the (land) Huang et al. (2016) emissivity database.	96
4.10	An example upwelling FSI spectrum simulated using RTTOVv12 overplotted with channels selected for (a) IASI and (b) FSI (FORUM) retrievals. The main diagonal of the measurement covariance S_y is shown in red in both cases. For IASI, S_y is derived from the differences between simulations and observations. For FSI, S_y is based on a combination of the target NESR and ARA and is the same as the green line in Figure 4.6a.	97
4.11	Example Jacobians of (a) temperature (K_T) and (b) water vapour (K_{WV}) for 7 of the 200 channels used in the retrieval. The Jacobians were calculated based on a mid-latitude profile (profile 42) in the ECMWF diverse-83 profile set using RTTOVv12. The temperature and water vapour (WV) profiles are shown in black. The Jacobians are given in terms of radiance units (RU), which are $\text{mW m}^{-2}\text{ sr}^{-1}/\text{cm}^{-1}$. For clarity, the Jacobian for water vapour has been scaled by the water vapour profile.	98
4.12	The median cumulative degrees of freedom for signal (CDOFS) for (a) water vapour (WV) and (b) temperature (T) averaged across all 240 cases for the IASI and FSI optimised configurations in red and blue, respectively. Shaded regions represent the median absolute deviation. This shows how the distribution of the vertical information can be related to the vertical resolution of the retrieved profile.	100

4.13	The median (a) water vapour (WV) and (b) temperature (T) bias of the retrieved profile to the 'true' state across the testing cases for the optimised IASI and FSI (FORUM) configurations in red and blue, respectively. Biases are evaluated between 300 and 1000 hPa in 100 hPa bins as in Trent et al. (2023). The water vapour percentage bias is calculated from absolute values in ppmv. Dashed lines show the median estimated standard deviation (ESD) for a single retrieval for the test cases in each bin. For water vapour, this is plotted with an offset of - 10% for clarity.	101
4.14	The median residual between the retrieved and 'true' (a) surface emissivity and (b) surface skin temperature for the IASI (red) and FSI (blue) configurations across the 240 cases. The shaded regions and error bars represent the median absolute deviation around this residual.	101
5.1	(a) The track of the C153 flight. (b) The altitude measured by the aircraft during the SLRs. Dropsonde releases are marked on (a) and correspond to the dashed lines in (b).	107
5.2	(a) The normalised standard deviation (%) of water vapour in ppmv (WV) and (b) standard deviation of temperature (T) taken from the ERA5 data surrounding the path of each dropsonde (± 30 mins, ± 48 km). The spatial resolution of ERA5 is $0.25 \times 0.25^\circ$ with a temporal resolution of one hour, and thus this plot gives an indication of the variability in the atmosphere at these scales.	109
5.3	(a)-(h) The FORUM-aircraft observations selected in Table 5.1 labelled with their corresponding dropsonde (top left). Observed radiances are given in radiance units (RU) which are $\text{mW}/\text{m}^{-2}/\text{sr}/\text{cm}^{-1}$. Each FORUM-aircraft observation is composed of one TAFTS and six ARIES scans, as described in Chapter 3. (i) The uncertainty (Unc.) in each FORUM-aircraft observation is shown in red, and is the noise, calibration (Figure 3.2b and c) uncertainty, and apodisation uncertainty (light blue in Figure 3.6) combined in quadrature. The variability (Var.) across all eight FORUM-aircraft observations is shown in blue and represents one standard deviation. The noise (yellow) of a single FORUM-aircraft observation.	111
5.4	A summary of the uncertainties associated with the retrievals from FORUM-aircraft observations for the C153 flight. The purple line is the residual between an FSI apodised LBLRTMv12.11 simulation and RTTOVv12 FSI simulation using the aircraft-level coefficients in the (a) TAFTS and (b) ARIES spectral ranges. The dashed line is the same as in Figure 5.3i, and is the noise (Figure 3.2b) and calibration (Figure 3.2c) uncertainty, and apodisation uncertainty (light blue in Figure 3.6) combined in quadrature.	113

5.5	The square-root-diagonal of the measurement covariance (equivalent to the FORUM-aircraft uncertainty (Figure 5.3i) overlaid on the FORUM-aircraft observation closest to dropsonde 8 (Figure 5.3h). This uncertainty is composed of the instrument uncertainty and the uncertainty that arises from the FORUM-aircraft apodisation process discussed in Section 3.3. The instrument uncertainty is the radiometric calibration and random noise for each instrument combined in quadrature for a single TAFTS scan and six ARIES scans. For both instruments, this uncertainty is dominated by the calibration, particularly for ARIES, where the random noise is reduced due to multiple scans (Figure 3.2b and c). The impact of the FORUM-aircraft apodisation process can be seen most clearly in the $15\ \mu\text{m}$ CO_2 band and between $1200\text{-}1400\ \text{cm}^{-1}$. The 200 channels selected for the aircraft retrievals are also shown in orange. . . .	114
5.6	The median (a) water vapour (WV) and (b) temperature (T) bias of the retrieved profile to the true state across the testing cases for the optimised FSI and FORUM-aircraft configurations in blue and orange respectively. Biases are evaluated between 300 and 1000 hPa in 100 hPa bins as in Trent et al. (2023). Dashed lines show the median estimated standard deviation (ESD) for a single retrieval for the test cases in each bin. For water vapour this is plotted with an offset of - 10% for clarity.	115
5.7	The median residual between the retrieved and input (a) surface emissivity and (b) surface skin temperature for the FSI and FORUM-aircraft configurations across the 240 cases in blue and orange, respectively. The shaded regions and error bars represent the median absolute deviation around this residual. The FSI emissivity residual shown here is the same as in Figure 4.14.	115
5.8	The retrieved surface emissivity in each of the 200 channels used in the retrievals from the eight spectra selected in Section 5.3. The shading represents one ESD in the retrieved value. The mean retrieved surface skin temperature was found to be $280.3 \pm 0.2\ \text{K}$	117
5.9	The mean retrieved (a) water vapour and (b) temperature profiles, as well as the (c) residual between the retrieved temperature and each AK-treated dropsonde profile (Drops_AK). Retrievals are shown using the global and local grid a-priori covariances for temperature in red and blue, respectively. The Drops_AK water profile and temperature profiles shown in (a) and (b) did not change significantly with the different covariances and so are shown only for the global covariance. The temperature residuals shown in (c) were calculated with respect to the correct Drops_AK profile. The global a-priori covariance is calculated from ERA5 temperatures across the globe for three days as used in Trent et al. (2023). The local a-priori covariance is calculated from ERA5 temperatures surrounding the SLRs for the duration of the flight. Blue and red shaded regions show the full spread of retrieved profiles with purple regions showing the overlap. The grey shaded region in (a) shows the full spread of AK-treated dropsonde profiles as well as the measurement uncertainty, and in (b) and (c) shows the dropsonde measurement uncertainty only.	117
5.10	The local a-priori covariance for temperature used in retrievals from aircraft observations. This was calculated using ERA5 data surrounding the SLR for the duration of the flight ($54.5\text{-}57.25^\circ\text{N}$ and $6.25\text{-}1.75^\circ\text{W}$ for 11:00-13:00 UTC).	118

5.11	Rows of the mean averaging kernels (AK) for (a,b) water vapour and (c,d) temperature calculated from retrievals of the C153 observations with colours representing the dropsonde altitude of each profile level. (a) and (c) follow the same colour scheme and are the rows of the AKs when the a-priori temperature covariance was derived from three days of global ERA5. (b) and (d) follow the same colour scheme and are the rows of the AKs when the a-priori temperature covariance was derived from the ERA5 surrounding the SLRs for the duration of the flight. The water vapour a-priori covariance used is the same in both sets of retrievals. The mean and standard deviation of the respective DOFS from the eight retrievals are shown for each plot.	120
5.12	(a) Water vapour (WV) retrievals and (b) the difference between the retrieved temperature and AK-treated dropsonde profiles (T Diff) performed on the FORUM-aircraft radiances. The shaded region in each retrieved value represents two ESD of the retrieval derived from the square-root diagonal of S_x . The a-priori is shown in orange, with the shaded region showing the a-priori standard deviation. The dropsonde profiles from each case are shown in red, and each one has had their respective averaging kernels applied (Drops_AK), which are shown in black. The shading around the dropsonde and Drops_AK profiles represents the uncertainties outlined in Section 5.2.1. For temperature, the dropsonde uncertainty is represented as 0.2 K. For water vapour, the dropsonde uncertainty varies with altitude but remains below 9%.	121
5.13	(a) The mean weighting function (change in transmission (Tr) with altitude (z)) for each wavenumber (WN) bin averaged across the 200 channels for all 8 dropsondes. (b) The radiance residual between the retrieved and FORUM-aircraft observed spectra for all 8 cases in each bin. Residuals have been averaged over 100 cm^{-1} bins, and the mean and standard deviation are shown for each bin. The black line represents the mean uncertainty (Unc.) from Figure 5.5, which is the square-root diagonal of S_y , in that bin. (c) The mean absolute improvement in the spectral residual from the first guess (FG) spectra to the retrieved spectra across all 8 cases in each bin. Error bars show the minimum and maximum values across the 8 cases in each bin. (d) The number of channels in each bin.	123
6.1	A schematic of the contribution of a layer to the upward radiance observed at an altitude where $\tau = \tau^*$. This is adapted from Matricardi (2005), and excludes the scattering of solar radiance.	131
6.2	The flight track (black) for the B895 flight that took place on 13 March 2015 plotted on the cloud optical thickness product from the MODIS Terra satellite overpass at 10:40 UTC. The first straight and level run (SLR 1) is shown in light blue, and the location of the selected radiance observation is marked with a blue circle (09:48:39 UTC). Dropsondes released during SLR 1 are marked with a cross. Approximately 50 minutes after the radiance observations were taken, the aircraft descended through the cloud, taking in-situ measurements following the light green path between 10:32 and 11:32 UTC.	136

6.3	(a) The FORUM-aircraft observation taken at 09:48:39 UTC during the first SLR. The spectral gaps correspond to the separation between the TAFTS shortwave and longwave channel, and the separation between the TAFTS shortwave and ARIES longwave channel. (b) The uncertainties associated with the FORUM-aircraft observation. The NESR (orange) corresponds to one TAFTS scan and six ARIES scans, and is based on the single scan NESR shown in Figure 3.3b. The ARA (blue) for TAFTS and ARIES are the same as in Figure 3.3c. The apodisation (Apod.) uncertainty (violet) is explored in Chapter 3 and is the same as the violet line in Figure 3.6. The radiance units are $\text{mW}/\text{m}^{-2}/\text{sr}^{-1}/\text{cm}^{-1}$ and are used for both suplots.	137
6.4	(a) The range-corrected lidar signal of the cloud measured at 09:48:35 UTC. (b) The lidar-derived extinction coefficient of the cloud measured at 09:48:35 UTC. The (c) temperature and (d) water vapour profiles measured by dropsonde 1 at 09:47:48 UTC. The uncertainty in the dropsonde measurements are described in Section 6.3 and are estimated to be 0.2 K and $\sim 10\%$ for temperature and water vapour, respectively. The edges of the cloud extent are marked in purple, and the altitudes at which in-situ measurements of cloud habit and PSDs have been characterised are shown with dashed orange lines (6, 6.8, 7, 7.3, 7.6, 8.0, and 8.3 km).	139
6.5	The (black) particle size distributions measured by a Cloud 2-DS probe during the B895 flight on 13 March 2015 that were successfully fitted with a bimodal Gaussian distribution.	141
6.6	The percentage difference between the original RTTOVv12 cloudy coefficients, and the ones built in this work for the (a) absorption and (b) scattering coefficients, as well as the (c) backscattering parameter (B Parameter). Differences are shown for the CER limits within the cloud optical property models ranging from (5-60 μm).	144
6.7	The residual between the FORUM-aircraft observation (Figure 6.3a) and the FORUM-aircraft simulation described in Section 6.4.1 given in (a) radiance (Rad) with radiance units (RU) of $\text{mW}/\text{m}^2/\text{sr}/\text{cm}^{-1}$ and (b) brightness temperature (BT). The shading represents the uncertainty in the FORUM-aircraft observation and is composed of the instrument noise, calibration uncertainty, and uncertainty introduced through the FORUM-aircraft apodisation added in quadrature. Both units are given to ease comparisons made throughout this Chapter.	146
6.8	The radiance residuals between the GHM and SC model (red) and the GHM and ASC model (black) are shown for the (a) LBLDIS and (b) RTTOVv12 models. The vertical profile of temperature and water vapour, and ice crystal properties are taken from Bantges et al. (2020), and are based on the profile in Figure 6.4 with a CER of 34 μm and a CTH of 9 km used in both models. A COT of 0.82 is used in LBLDIS and an IWP of 28 gm^{-2} is used in RTTOVv12. The FSI target uncertainty for this case is shown in blue. The uncertainty associated with the FORUM-aircraft observation is shown in grey, and is composed of the instrument noise, calibration uncertainty, and the uncertainty associated with the FORUM apodisation process described in Chapter 3 added in quadrature. Each of these components were presented in Figure 6.3b.	147

6.9	The square-root of S_y used in the retrievals in radiance units. This is a combination of the instrument NESR, ARA, and uncertainty introduced through the FORUM-aircraft apodisation process shown individually in Figure 6.3b. . . .	150
6.10	(a) The channels selected for retrievals using the FORUM-aircraft configuration of IMS on top of the FORUM-aircraft simulation in brightness temperature (BT), with (b) the number of channels in each 50 cm^{-1} bin. The 200 channels selected in Chapter 5 for optimised retrievals of temperature and water vapour are shown in purple. The additional 26 channels that have been selected to optimise retrievals of ice cloud properties are shown in blue.	151
6.11	The brightness temperature (BT) residuals between the RTTOVv12 FSI simulated spectra assuming the GHM and SC databases for example CER values and an ice water path of $\sim 28 \text{ gm}^{-2}$ derived from the lidar measurements. The remainder of the atmospheric and surface profile are as described in Section 6.4.1. The uncertainty is square-root-diagonal of the measurement covariance in Figure 6.9, and is equivalent to the FORUM-aircraft uncertainty.	152
6.12	The BT residuals between the observed and retrieved spectrum and J_y in the cloud-sensitive channels using the (dashed) mid-infrared (MIR) only and (solid) mid- and far-infrared (MIR+FIR) on the FORUM-aircraft (a) LBLDIS simulation and (b) observation. The residuals within the mid-infrared are partially obscured by the combined MIR+FIR retrieval. The retrievals using the SC and GHM models are shown in green and purple, respectively. The square-root-diagonal of S_y is shaded in grey as the uncertainty.	154
6.13	The BT residuals between the observed and retrieved spectrum and J_y for retrievals from the observation. All 226 channels are used in the retrieval, but only residuals and J_y in the 26 cloud sensitive channels are shown for comparison to Figure 6.12. The square-root-diagonal of S_y is shaded in grey as the uncertainty. J_y across all 116 (MIR) and 226 (MIR+FIR) channels is as follows: SC (MIR): 0.33. GHM (MIR) 0.34. SC (MIR+FIR): 1.20. GHM (MIR+FIR): 1.15.	158
6.14	The retrieved (a) temperature and (c) water vapour profile from the FORUM-aircraft observation with (b) differences between the retrieved and AK-treated dropsonde temperature profiles. The retrievals assuming the GHM and SC models are shown in purple and green, respectively, and use all 226 channels. The average cloud base from the two retrievals is shown with the dashed purple line. The shaded region around each retrieval represents one ESD. The a-priori is shown in orange, with the surrounding shading showing one standard deviation. The AK-treated dropsonde (Drops_AK) profiles are shown in black. The uncertainty in the dropsonde measurements are discussed in Section 6.3 but are not visible on the scale of this figure. For temperature, the dropsonde uncertainty is 0.2 K. For water vapour, the dropsonde uncertainty varies with altitude but remains below 9%.	160

6.15 The retrieved (a) COT and (b) CER from the FORUM-aircraft observation in comparison to measurements of the cloud. The retrieved values are shown using squares (116 MIR channels) and triangles (226 MIR+FIR channels). The retrievals using the SC and GHM models are shown in green and purple, respectively. Error bars show one ESD in the retrieved CTH, COT, or CER. The measurements of the cloud taken by instruments on board the plane are shown as black dots. For the COT, the mean value derived from the lidar measurements between from 09:48:09 to 09:49:09 UTC is shown, with the error bar representing the standard error of the mean. For the CER, the black dots are calculated from the PSDs measured by the Cloud 2-DS probe at 5 altitudes within the cloud, and the error bars show one standard deviation derived from the bimodal fit. The vertical black line shows the median MODIS Level 2 COT and CER from the 10:40 UTC overpass between 58.7-59.75°N and 4.2-3.3°W at 1 km resolution. The shading around the MODIS COT and CER is one median absolute deviation and shows the spread of values within this spatial region. The a-priori for the CER is shown in orange with shading representing one standard deviation. The a-priori for the COT is 0.01 and is not shown. . . 162

List of Abbreviations

3V-CPI	3-View Cloud Particle Imager.
AERI	Atmospheric Emitted Radiance Interferometer.
AK	Averaging Kernels.
AMSU	Advanced Microwave Sounding Unit.
ARA	Absolute Radiometric Accuracy.
ARIES	Airborne Research Interferometer Evaluation System.
ASC	Aggregate Solid Columns.
AVAPS	Airborne Vertical Atmospheric Profiling System.
BT	Brightness Temperature.
CAM	Complex Angular Momentum.
CAMS	Copernicus Atmosphere Monitoring Service.
CDOFS	Cumulative Degrees of Freedom for Signal.
CER	Cloud Effective Radius.
CIP 100	Cloud Imaging Probe 100.
CIRCCREX	CIRrus Coupled Cloud-Radiation EXperiment.
COT	Cloud Optical Thickness.
CTH	Cloud Top Height.
DISORT	DIScrete-Ordinate Radiative Transfer Model.
DOFN	Degrees of Freedom for Noise.
DOFS	Degrees of Freedom for Signal.
ECMWF	European Centre for Medium-Range Weather Forecasts.
EEI	Earth's energy imbalance.
ERB	Earth's Radiation Budget.
ESA	European Space Agency.
ESD	Estimated Standard Deviation.
FAAM	Facility for Atmospheric Airborne Measurements.
FEI	FORUM Embedded Imager.
FINESSE	Far-INfrarEd Spectrometer for Surface Emissivity.
FIRMOS	Far-Infrared Radiation Mobile Observation System.
FIRR2	Far-InfraRed Radiometer-2.
FORUM	Far-infrared Outgoing Radiation Understanding and Monitoring.
FSI	FORUM Sounding Instrument.
FWHM	Full Width Half Maximum.
GHM	General Habit Mix.
HiSRAMS	High Spectral Resolution Airborne Microwave Sounder.
HITRAN	High-resolution Transmission Molecular Absorption.
IASI	Infrared Atmospheric Sounding Interferometer.
ILS	Instrument Line Shape.

IMS	Infrared Microwave Sounding.
INP	Ice Nucleating Particle.
IPCC	Intergovernmental Panel on Climate Change.
IWC	Ice Water Content.
IWP	Ice Water Path.
LBLDIS	LBLRTM+DISORT.
LBLRTM	Line-by-Line Radiative Transfer Model.
MERRA-2	Modern-Era Retrospective analysis for Research and Applications v2.
MHS	Microwave Humidity Sounder.
MODIS	Moderate Resolution Imaging Spectroradiometer.
MT_CKD	Mlawer-Tobin-Clough-Kneizys-Davies Water Vapor Continuum Model.
NESR	Noise-Equivalent-Spectral-Radiance.
NOAA	National Oceanic and Atmospheric Administration.
OLR	Outgoing Longwave Radiation.
OPD	Optical Path Difference.
PDF	Probability Density Function.
PREFIRE	Polar Radiant Energy in the Far Infrared Experiment.
PSD	Particle Size Distribution.
RAL	Rutherford Appleton Laboratory.
RMSE	Root-Mean-Squared-Error.
RT	Radiative Transfer.
RTTOVv12	Radiative Transfer for the TIROS Operational Vertical Sounder v12.
SC	Solid Columns.
SLR	Straight Level Run.
TAFTS	Tropospheric Airborne Fourier Transform Spectrometer.
TCWV	Total Column Water Vapour.
TOA	Top-of-Atmosphere.
UNIRAS	UNiversal InfraRed Airborne Spectrometer.
UWIREMIS	University of Wisconsin InfraRed land surface EMISsivity.
WHAFFERS	W-band, HiSRAMS, AERI, FIRR2, FINESSE and FIRMOS Experiment on Remote Sensing.

Chapter 1

Introduction

This thesis outlines the extension of the Rutherford Appleton Laboratory (RAL) Infrared Microwave Sounding (IMS) retrieval scheme in preparation for the European Space Agency's (ESA) Far-Infrared Outgoing Radiation Understanding and Monitoring (FORUM) mission. The FORUM satellite will be the first satellite to measure spectrally resolved far-infrared ($100\text{--}667\text{ cm}^{-1}$) radiances from the top of the atmosphere (TOA) at a high spectral resolution and is due to launch in 2027. The observations from FORUM are expected to provide significant information about the distributions of water vapour and the properties of ice clouds (Palchetti et al. 2020).

This introduction will explore key concepts underpinning the work in this thesis. Section 1.1 describes the Earth's Radiation Budget and the significance of water vapour and ice clouds. Section 2.1 outlines the theory of radiative transfer. Section 1.2 describes the far-infrared and the ESA FORUM mission. Section 1.3 describes the motivation for this thesis, and the overall structure of the thesis is presented in Section 1.4.

1.1 Earth's Radiation Budget

The Earth's Radiation Budget (ERB) summarises the TOA balance between incoming solar radiation ($0.1\text{--}4\text{ }\mu\text{m}$) and the loss of radiation to space. A summary of the radiative fluxes associated with the Earth's globally averaged radiation budget from 2006-2018 is presented in Figure 1.1 (Forster et al. 2021).

Figure 1.1 highlights that the loss of radiation to space is primarily a combination of two effects. The first is the solar albedo effect, which represents the reflection of solar radiation back to space, creating a cooling response on the Earth's atmosphere and surface. The second is the infrared greenhouse effect, which occurs when the upwelling thermal radiation emitted by the Earth's surface is partially absorbed and re-emitted by atmospheric components, trapping the radiation within the Earth's atmosphere. This creates a warming response on the Earth's atmosphere (Liou 2002). The contribution of these two effects to the loss of radiation to space is dictated by the nature of the Earth's atmosphere and surface.

The Intergovernmental Panel on Climate Change (IPCC) estimated that the ERB for the period between 2006-2018 had a net imbalance of $0.7 [0.5-0.9] \text{ Wm}^{-2}$. This imbalance is referred to as the Earth's energy imbalance (EEI), and creates a positive forcing and warming response on the Earth's atmosphere and surface (Forster et al. 2021). Satellite

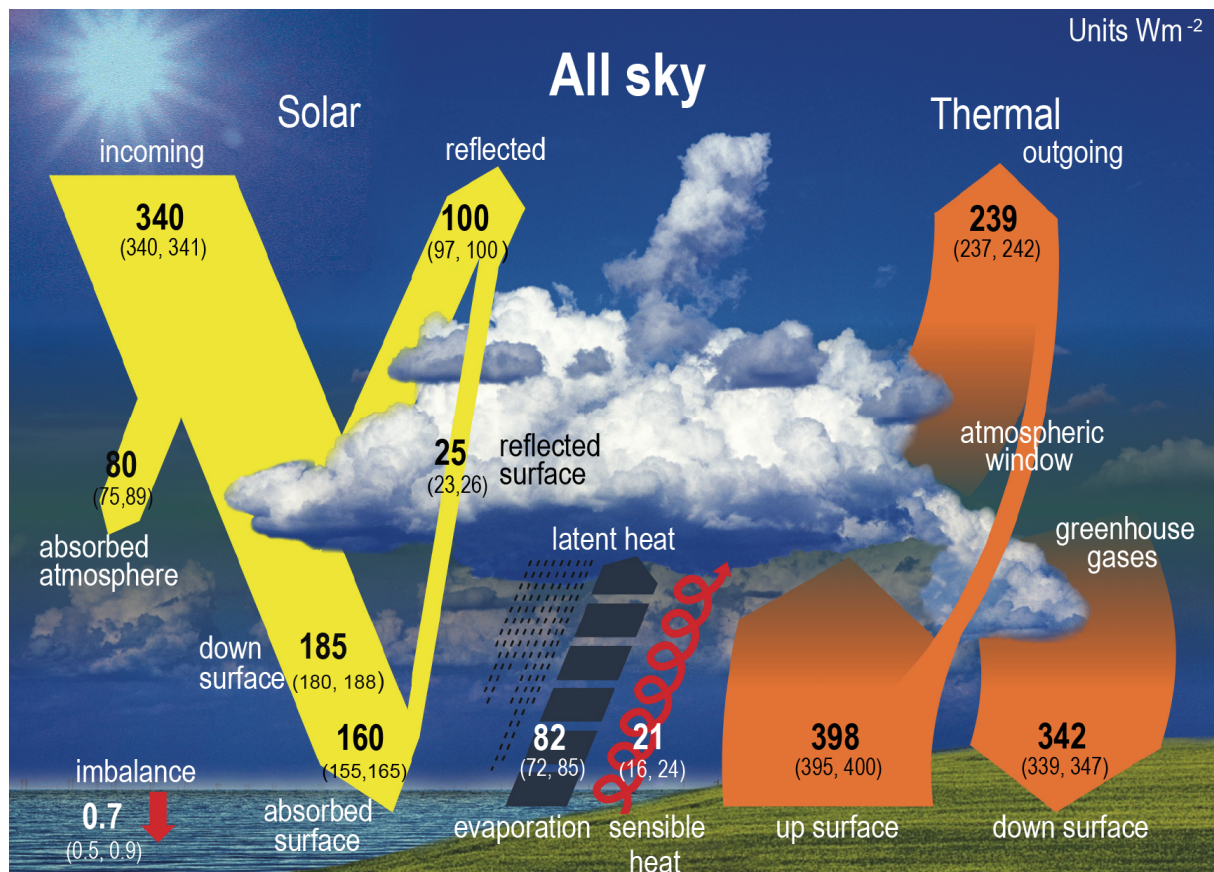


Figure 1.1: The radiative fluxes associated with the Earth's globally averaged radiation budget from 2006-2018. Numbers indicate the best estimates for the magnitudes in Wm^{-2} , with uncertainties shown in parentheses (5-95 % confidence range). Taken from Forster et al. (2021)

observations show that the EEI is due to increase, with anthropogenic activities trapping more of the outgoing thermal radiation (Raghuraman et al. 2021). Properly constraining the EEI is therefore vital as an indicator and measure of anthropogenic climate change (von Schuckmann et al. 2023). There are a number of techniques that are currently used to constrain the EEI. These include measurements of the EEI itself using TOA radiometric observations (Loeb et al. 2024), satellite measurements of sea level and ocean mass change (Meyssignac et al. 2023, Hakuba et al. 2024), and an in-situ based Earth heat inventory that quantifies and locates where the heat has accumulated in the Earth system (von Schuckmann et al. 2023).

Most of the additional heat caused by the EEI is stored in the ocean (89 %), with the rest divided between the land (6 %), cryosphere (4 %), and atmosphere (1 %) (von Schuckmann et al. 2023). This has significant repercussions on the systems contained within each sphere. The warming of the ocean not only impacts core marine ecosystems (Wernberg et al. 2024), but contributes towards an increase in the global mean sea level through ocean heat expansion (Hakuba et al. 2024). The additional heat stored in the land leads to an increase in ground surface temperatures, which impacts the storage of carbon and water in the soil affecting vegetation and crops (Cuesta-Valero et al. 2021, Mu et al. 2021). The increasing temperatures in the cryosphere have led to the thawing of permafrost which has increased greenhouse gas concentrations in the atmosphere (Turetsky et al. 2020), and significant sea ice, ice sheet, and glacier loss which impacts the global mean sea level, ocean-atmosphere circulations, and reduces the reflection of solar radiation (Cohen et al. 2019, Slater et al. 2021). Finally, the heating of the atmosphere leads to an increase in air temperature that, most significantly for this work, impacts the tropospheric water vapour content and formation of clouds (Forster et al. 2021)

The remainder of this section will discuss the role of water vapour (Section 1.1.1) and cirrus clouds (Section 1.1.2) within the ERB and EEI in more detail, particularly focusing on their participation in the infrared greenhouse effect.

1.1.1 The Role of Water Vapour

Greenhouse gases cause a significant absorption of the outgoing thermal radiation, trapping heat within our atmosphere. Water vapour composes approximately 3% of the Earth's atmosphere and is the most dominant greenhouse gas with absorption bands in the microwave, infrared, and visible spectral regions. It can reach concentrations of up to the order of 10^5 ppmv closer to the surface, and reduce to as low as 1 ppmv closer to the tropopause and lower stratosphere as a result of the exponential dependence of saturation vapour pressure on air temperature (Andrews 2010). It also plays a vital role in the Earth's hydrological cycle, and in the Earth's energy cycle, with the latent heat of water vapour accounting for approximately half of the poleward, and most of the upward, heat transport within the atmosphere (Allan 2011, Sherwood et al. 2010). Therefore, capturing its spatial and temporal distribution is critical in estimating the strength of water vapour's role in the ERB (Dessler et al. 2008).

In addition, the water vapour feedback is the strongest positive feedback in the climate system (Forster et al. 2021). An increase in the atmospheric temperature increases the saturation vapour pressure, which means that the air can hold more water vapour. More water vapour can trap more of the outgoing thermal radiation, which increases atmospheric temperatures and forms a strong positive feedback (Colman & Soden 2021). This feedback can be triggered by an increase in other greenhouse gases, and is also clearly tied to the formation of clouds as it intensifies the global water cycle (Forster et al. 2021).

Water vapour in the upper troposphere strongly regulates this water vapour feedback (Chung et al. 2014), and it has been shown that it is likely that human influence has contributed to the current increase in upper tropospheric moisture, amplifying the water vapour feedback (IPCC 2023). However, discrepancies exist in long-term trends of upper tropospheric water vapour between observations, climate models, and reanalysis datasets (Schröder et al. 2019, Santer et al. 2021, Allan et al. 2022, Trent et al. 2024). Considerable work has been performed to improve assessments of water vapour distributions through the use of in-situ measurements such as radiosondes and aircraft-based sensors (Sun et al. 2021). However, best-case uncertainties from the GCOS Reference Upper-Air Network radiosondes are estimated to still be of the order 5 % in relative humidity, and reach 15 % near the tropopause

(Dirksen et al. 2014). There is also a limited homogeneity in the radiosondes' distribution, regularity, and vertical sampling (Ferreira et al. 2019). Similarly, while frost point hygrometers have a greater sensitivity to upper tropospheric and lower stratospheric water vapour, cryogenic frost point hygrometers have uncertainties of at least 5 % in relative humidity (Vömel et al. 2016), and the National Oceanic and Atmospheric Administration (NOAA) frost point hygrometer has an uncertainty of up to 12 % in mass mixing ratio in the troposphere (Hall et al. 2016).

1.1.2 The Role of Cirrus Clouds

Cirrus clouds are high-altitude ice clouds that can form under a variety of conditions and cover ~ 30 % of the Earth (Stubenrauch et al. 2006). At temperatures below 235 K and a relative humidity with respect to ice above 140 %, pure water droplets can freeze into ice crystals through homogeneous nucleation. This can occur when a moist air parcel ascends to higher altitudes where there are colder temperatures. If these conditions are not met, heterogeneous nucleation occurs if an ice nucleating particle (INP), such as an aerosol, is present to trigger the freezing of water droplets or the deposition of atmospheric water vapour (Lohmann et al. 2016). The size, surface composition, and morphology of this INP can lead to different submechanisms and, thus, variations in the size, shape (referred to as the habit) and distribution of the ice crystals within the cloud (Zhao et al. 2019).

The net radiative effect of a cirrus cloud reflects a competition between the solar albedo and infrared greenhouse effect and is dependent on their coverage, position, geometric thickness and microphysics (Yang et al. 2005). The infrared greenhouse effect occurs for cirrus clouds, as they can absorb or scatter the upwelling thermal radiation. A partial absorption reduces the energy lost to space and allows the cloud to radiate energy at longer wavelengths, associated with the lower temperatures at its altitude (Lohmann et al. 2016). There is strong evidence suggesting that cirrus clouds have a net warming effect on the atmosphere through enhanced trapping of the outgoing thermal radiation (Sassen et al. 2008). This is particularly true for high cirrus clouds that are also transparent to the downwelling solar radiation, but opaque to the outgoing thermal radiation (Hong et al. 2016).

The cloud feedback is the coupling between cloudiness and surface air temperature, with an increased surface temperature leading to changes in the cloud macro- and microphysics that either amplify or diminish the initial temperature perturbation (Lohmann et al. 2016). Studies suggest that interannual surface warming driven by anthropogenic activities is increasing the number of a specific type of cirrus clouds that have a net warming effect on the atmosphere (Haywood et al. 2009, Zhou et al. 2014, Zhao et al. 2019, Singh et al. 2024).

There are currently large disparities between climate model predictions of the net radiative forcing and feedback processes of cirrus clouds (Forster et al. 2021). This is in part due to a limited understanding of their formation and development, as well as their variation in ice crystal microphysics (Fu et al. 2017). In comparison to water clouds, the variation in potential ice crystal habits introduces additional complexity for calculating their radiative interactions (Baran et al. 2014). Poorly constraining these habits can also result in significant errors in our understanding of the other cloud microphysical properties (Mishchenko et al. 1996, Yang et al. 2015).

1.2 The Far-Infrared

The values of radiative flux shown in Figure 1.1 are averaged globally and temporally and integrated spectrally. Over 99% of the outgoing thermal radiation is in the spectral range $100\text{-}2500\text{ cm}^{-1}$ ($4\text{-}100\text{ }\mu\text{m}$) and is called the Outgoing Longwave Radiation (OLR).

An example of the spectrally resolved OLR is shown in Figure 1.2 in terms of radiance (I) for a tropical atmosphere in clear-sky and cloudy conditions. The radiance is the energy transferred by radiation passing through a unit solid area, in a given direction, per unit of time, at a specific wavenumber (inverse wavelength with units cm^{-1}) and is commonly measured in $\text{mWm}^{-2}\text{sr}^{-1}/\text{cm}^{-1}$.

In the clear-sky case, the fine spectral features shown in Figure 1.2 are a result of the absorption and emission of gases within the Earth's atmosphere that attenuate the radiation reaching the TOA (Andrews 2010). The inclusion of a cloud in the blue curve can be seen to reduce the amount of thermal radiation reaching the TOA between $\sim 200\text{-}1500\text{ cm}^{-1}$. The

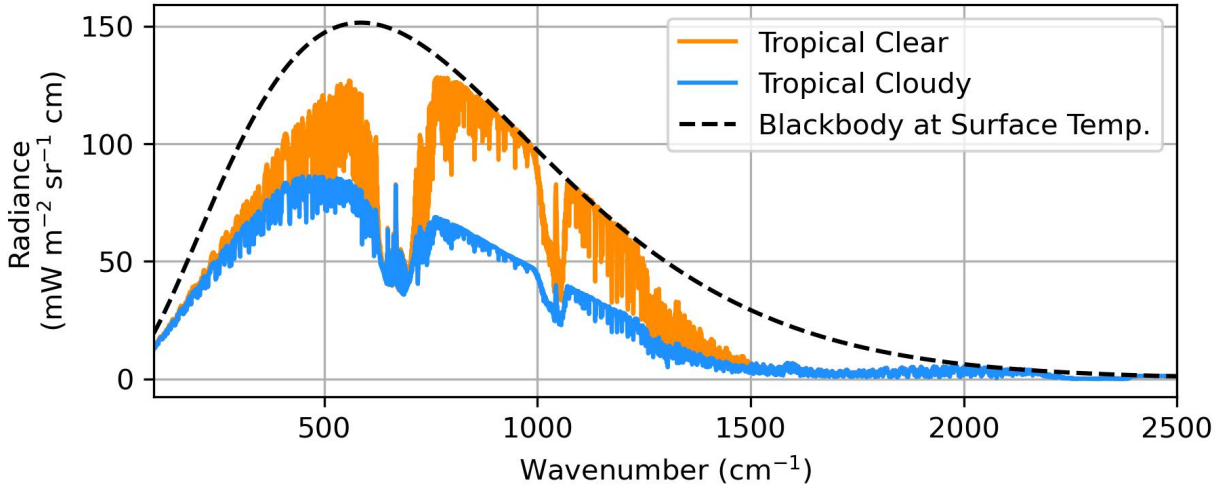


Figure 1.2: Example spectra simulated using a radiative transfer code for a tropical profile in a clear sky (orange) and with an example ice cloud in the upper troposphere (blue) atmosphere. The blackbody curve for the surface temperature is shown in black.

extent of this impact is dependent on the properties of the ice cloud, and does also impact the finer structures observed throughout the OLR spectrum. This thesis exploits the spectral features observed in the OLR spectrum that are caused by the transfer of radiation through the atmosphere, and this will be described in more detail in Chapter 2.

Theoretical models suggest that globally, approximately half of the OLR is within the far-infrared region ($100\text{--}667\text{ cm}^{-1}$) (Palchetti et al. 2020). Despite this, the OLR spectrum across the far-infrared has only been measured for the first time last year (L’Ecuyer et al. 2021). Recent developments in uncooled detectors that are sensitive to far-infrared radiances have enabled new space missions with enhanced accuracy and coverage to be proposed, promoting further research from ground or airborne instruments in their support. This section will briefly describe instruments that have measured spectrally resolved radiances in the far-infrared before describing the FORUM mission and the motivation for this thesis.

1.2.1 Instruments In The Far-Infrared

Currently, very few instruments can observe spectrally resolved atmospheric radiances in the far-infrared and only one at present from the TOA. Below is a list of instruments that have observed or currently observe spectrally resolved radiances in the far-infrared. Key details about their spectral ranges, spectral resolutions, and viewing formations are summarised in

Table 1.1: A summary of the spectral ranges and resolution for each instrument as well as their measurement of the upwelling or downwelling radiation (Mlynchak et al. 2006, Bianchini et al. 2019, Canas et al. 1997, Palchetti et al. 2021, Murray et al. 2024, Hanel et al. 1970, 1971, Kempe et al. 1980, L’Ecuyer et al. 2021). The spectral resolution for PREFIRE is specified in wavelength.

Instrument	Spectral Range (cm^{-1})	Spectral Resolution (cm^{-1})	Measurement and Position
FIRST	50-2000	0.625	Upwelling ; Balloon Flight
REFIR-PAD	100-1900	0.4	Upwelling; Balloon Flight Downwelling; Ground
TAFTS	80-600	0.12	Upwelling ; Airborne
FIRMOS	100-1700	0.4	Downwelling ; Ground
FINESSE	400-1600	0.5	Downwelling ; Ground
IRIS-B	400-2000	5	Upwelling ; Satellite
IRIS-D	400-1600	2.8	Upwelling ; Satellite
SI	400-1600	5	Upwelling ; Satellite
PREFIRE	185-2000	($0.86 \mu\text{m}$)	Upwelling ; Satellite

Table 1.1. Some of these radiances have been used in support of the FORUM mission (Section 1.2.2) and to perform the first retrievals of water vapour and ice cloud properties (Section 1.3) that provide the foundation for this work.

NASA’s Far InfraRed Spectroscopy of the Troposphere (FIRST) instrument has been used for a number of balloon and ground-based campaigns, such as the RHUBC-II campaign in 2017, which aimed to improve the modelling of water vapour spectroscopy (Mlynchak et al. 2006, Mast et al. 2017).

Radiation Explorer in the Far-Infrared - Prototype of Applications and Development (REFIR-PAD) has been used on stratospheric balloon platforms to measure upwelling radiances (Bianchini et al. 2019), as well as on the ground to measure downwelling radiances and studies of the water vapour continuum absorption in the far-infrared (Serio et al. 2008). The instrument is currently installed at Concordia base at Dome-C where it has been operating since 2012 and has contributed to a number of studies of the far-infrared (Maestri et al. 2014, Di Natale et al. 2017, Maestri et al. 2019, Di Natale et al. 2020)

Tropospheric Airborne Fourier Transform Spectrometer (TAFTS) is the only instrument

to have measured spectrally resolved upwelling far-infrared radiances from an aircraft. It has been used in studies of surface emissivity (Bellisario et al. 2017, Murray et al. 2020), clear sky and ice clouds (Warwick et al. 2022, Cox et al. 2007, 2010), and the water vapour continuum (Fox et al. 2015, Green et al. 2012). A more detailed description of TAFTS is contained within Chapter 3, and its observations are used throughout this thesis.

Far-Infrared Mobile Observation System (FIRMOS) was modelled on REFIR-PAD and was developed for ground-based measurements (Palchetti et al. 2021). It was situated at Zugspitze Summit to carry out a two-month campaign for both clear-sky and ice cloud acquisitions similar to REFIR-PAD (Belotti et al. 2023). It also participated in the W-band, HiSRAMS, AERI, FIRR2, FINESSE and FIRMOS Experiment on Remote Sensing (FIRMOS) campaign that took place in 2025 in Northern Canada, observing downwelling radiances in both clear and cloudy conditions (Brindley et al. 2025).

Far-INfrarEd Spectrometer for Surface Emissivity (FINESSE) is a ground-based instrument that has a rotating mirror enabling it to take measurements from nadir to zenith viewing angles (Murray et al. 2024). Its primary use is to take measurements of the spectral surface emissivity in the far-infrared (Warwick et al. 2024), but it has also been used to take measurements of downwelling radiances. To date, it has participated in two campaigns: the Mixed Phase Clouds and Climate campaign, which took place in 2022 in Andøya, Norway, and the WHAFFFFERS campaign, which took place in 2025 (Brindley et al. 2025).

The Infrared Interferometer Spectrometer (IRIS) experiment included two satellite-based instruments, IRIS-B and IRIS-D, that were placed on the NASA Nimbus 3 and Nimbus 4, respectively. These satellites launched in 1969 and 1970, respectively (Hanel et al. 1970, 1971). The aim of this experiment was to provide information on the vertical structure of the atmosphere, particularly carbon dioxide and water, as well as the Earth's surface emissivity.

The Spektrometer Interferometer (SI) was an instrument on board the METEOR-25 and METEOR-28 satellites that launched in 1977 and 1979, respectively. These also aimed to provide information on the vertical structure of the atmosphere, particularly carbon dioxide (Kempe et al. 1980).

Polar Radiant Energy in the Far Infrared Experiment (PREFIRE) is a NASA Earth Venture

Instrument which aims to measure far-infrared signatures of the atmospheric and surface processes in the Arctic and Antarctic regions from space. It consists of two CubeSats that each cover 64 spectral channels and was launched in May and June 2025 with a nominal lifetime of 1 year (L'Ecuyer et al. 2021).

1.2.2 The Far-Infrared Outgoing Radiation Understanding and Monitoring Mission

This study is in support of the ESA FORUM mission, which aims to measure the Earth's OLR in the far-infrared with a high accuracy and spectral resolution. These measurements will be used to target three scientific areas. The first is to assess and improve the underlying spectroscopy in the far-infrared. The second is to link the observed far-infrared signatures to the atmospheric variability of, in particular, water vapour, greenhouse gases, and clouds, as well as to the surface properties. The third is to use these observations to evaluate the radiative processes within climate models (Oetjen et al. 2020).

The FORUM mission consists of two instruments. The primary instrument is the FORUM Sounding Instrument (FSI), which will have a spectral range from $100\text{--}1600\text{ cm}^{-1}$ with a spectral resolution greater than 0.5 cm^{-1} and a target radiometric accuracy of 0.1 K at 3σ (Palchetti et al. 2020). This instrument has a ground footprint of 15 km , based on an orbital altitude of 830 km , and uses the stop-and-stare technique to view the scene (Oetjen et al. 2020). The second instrument is the FORUM Embedded Imager (FEI), which is a standard imager co-aligned with the FSI that operates at $10.5\text{ }\mu\text{m}$ to identify clouds and subpixel heterogeneities in the observed scene, and has a ground footprint of 36 km that surrounds the FSI footprint (Figure 1.3).

The FORUM satellite will fly in a loose formation with the EUMETSAT MetOp-SG-1A satellite, complementing observations of the OLR taken by the Infrared Atmospheric Sounding Instrument New Generation (IASI-NG) that cover $647\text{--}2760\text{ cm}^{-1}$ with a spectral resolution of 0.25 cm^{-1} (Figure 1.3). When combined, these two instruments will create a unique dataset of the Earth's entire OLR spectrum (Palchetti et al. 2020).

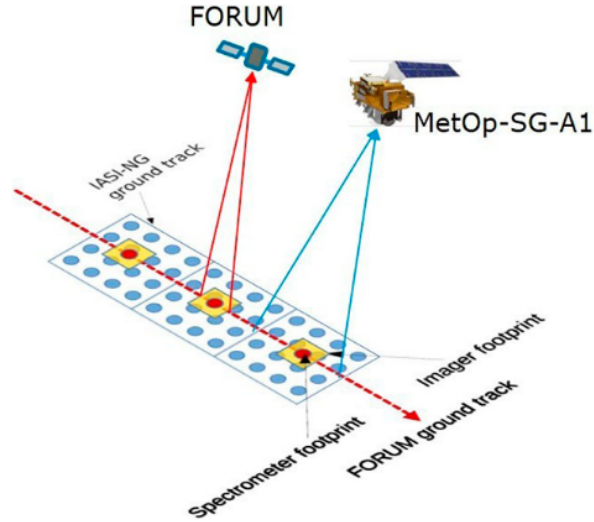


Figure 1.3: The satellite ground tracks and footprints for the FSI (red), FEI (yellow), and IASI-NG (blue). Taken from Palchetti et al. (2020)

1.3 Exploiting the Far-Infrared in Retrievals

In the field of remote sensing, retrievals are used to infer information about a physical parameter of Earth from measurements of emitted or scattered radiation taken from the ground, an airborne platform, or a satellite. The more sensitive the measurement of radiation to the physical parameter, the more information is available about it. Radiances observed in different spectral regions exhibit different sensitivities to retrieval targets, and as a result, satellite observations taken across the electromagnetic spectrum have been used to characterise the atmosphere. These observations can be taken by either active instruments, which generate radiation such as microwaves or radar to sound the atmosphere, or passive instruments, which measure the radiation that is naturally emitted, such as the OLR, and are advantageous as they have a large spatial coverage (Liou 2002).

Currently, satellite retrievals of water vapour can lack sufficient vertical resolution (Chung et al. 2014), demonstrate an inherent bias (Santer et al. 2021), or have a reduced sensitivity to upper tropospheric water vapour (Kursinski & Gebhardt 2014), causing inconsistencies between retrieved upper tropospheric humidities (Shi et al. 2022). While hyperspectral sounders such as Atmospheric Infrared Sounder (AIRS) and the Infrared Atmospheric Sounding Interferometer (IASI) have improved the vertical resolution of water vapour and temperature (Chahine et al. 2006, Hilton et al. 2012), radiances in the mid-infrared ($667\text{--}2000\text{ cm}^{-1}$) have

a limited sensitivity to water vapour in the mid- to upper troposphere, and biases in the upper tropospheric remain present (Fetzer et al. 2008, Trent et al. 2019). Similarly, using passive remote sensors that cover the mid- and near-infrared for ice cloud retrievals has limitations due to the spectral contributions of surface reflectance, particularly over land, for optically thin cirrus (Baran et al. 1998, Chepfer et al. 2002). Active sensors have also been used to estimate ice crystal habits from observations. However, these observations have a low spatial coverage and are limited by the geometric thickness of the cloud (Saito et al. 2017, Pfreundschuh et al. 2022).

The absorption of water vapour dominates the far-infrared. As a consequence, the OLR in this region is significantly more sensitive to mid- and upper tropospheric water vapour than in the mid-infrared (Sinha & Harries 1995, Brindley & Harries 1998). Theoretical studies by Merrelli & Turner (2012) have demonstrated this potential by performing retrievals of simulated upwelling radiances with realistic instrument configurations. Furthermore, the benefit of including far-infrared information has recently been confirmed by Warwick et al. (2022) who performed a single retrieval on a spectrum covering the mid- and far-infrared observed from an aircraft flying in the upper troposphere. To date, other existing retrieval codes built to exploit the far-infrared signals of water vapour have only been tested on simulations of upwelling far and mid-infrared radiances, and observations of downwelling radiances limited to the spectral range $200\text{-}1000\text{ cm}^{-1}$ (DiNatale et al. 2020, Ridolfi et al. 2020).

Similarly, features of the refractive index of ice within the far-infrared mean it could be advantageous to include far-infrared wavenumbers in retrievals of ice cloud properties. The real component of the refractive index of ice (related to scattering) is much larger than the imaginary component (related to absorption) within the far-infrared at $\sim 410\text{ cm}^{-1}$. This suggests significant scattering would occur in this region that would imply a sensitivity to ice crystal habit (Warren & Brandt 2008), which studies suggest could be detected by the FSI (Palchetti et al. 2020, Bantges et al. 2020, DiNatale & Palchetti 2022). Furthermore, simulation studies have indicated that including a few far-infrared channels in current retrievals of ice cloud properties from existing mid-infrared spaceborne radiometers could significantly reduce the uncertainties of COT, CER, and CTH, particularly in polar regions and the upper

tropical troposphere (Libois & Blanchet 2017). This is in part due to the larger variations between the refractive index of ice and water in the far-infrared than in the mid-infrared, making it easier to separate spectral contributions of ice clouds and water vapour (Warren & Brandt 2008, Libois & Blanchet 2017).

Some retrievals have also been performed on observations of downwelling far-infrared radiances in the presence of ice clouds. However, these were limited by uncertainties in the water vapour and temperature profiles that would be less dominant when looking at upwelling radiances (Di Natale et al. 2017, Maestri et al. 2014). At present, there are no published retrievals from observed upwelling far-infrared radiances in the presence of a cirrus cloud, and only two published studies have attempted to replicate upwelling far-infrared radiances measured under these conditions. Cox et al. (2010) was limited by large uncertainties in the atmospheric state and a lack of instrumentation capable of measuring small ice particles. Bantges et al. (2020) found that current ice cloud optical property models could not simultaneously simulate mid- and far-infrared radiances within the instrument uncertainty. Despite these findings, these optical property models are still commonly used to characterise ice clouds from far-infrared radiances (Magurno et al. 2020, Di Natale & Palchetti 2022, Peterson et al. 2022).

Given the sensitivities and spectral features across the far-infrared for water vapour and ice clouds, there is a significant potential for better retrievals of water vapour and ice cloud properties using the FORUM observations (Ridolfi et al. 2020). The work in this thesis aims to develop a retrieval scheme that exploits this potential and can be applied to observations from the FSI following the FORUM launch. Within this thesis, this retrieval scheme has been tested on airborne observations of upwelling far- and mid-infrared radiances exploiting the full FSI spectral range, and is the only retrieval scheme to have been used in this way.

1.4 Structure of Thesis

The aim of this thesis is to adapt the IMS retrieval scheme to be available for use on FORUM observations. It has undergone three stages of testing to prepare it. The first stage includes

testing on a diverse set of simulations. The second stage contains an application to upwelling far-infrared radiances observed from an aircraft in clear-sky conditions. The final stage describes the first retrieval of ice cloud properties from an airborne observation of upwelling far-infrared radiances above a cirrus cloud layer. The content of each chapter in this thesis is as follows:

- Chapter 2 of this thesis outlines the theory of radiative transfer and the mathematical basis of the retrieval method and common diagnostic parameters used to evaluate the final retrievals.
- Chapter 3 introduces the theory of Fourier transform spectroscopy and details the instruments used to collect radiances. It also describes how observed radiances have been adapted to mimic the FSI spectral characteristics.
- Chapter 4 details the RAL IMS retrieval scheme in its previous form, before outlining how it has been extended for the FSI. It also includes information on testing on a variety of TOA spectral simulations. Chapter 5 includes the testing of IMS on radiance observations taken during the PIKNMIX-F airborne campaign by Jon Murray and Laura Warwick, and an evaluation against in-situ measurements. Elements of Chapters 3-5 have been published in Panditharatne et al. (2025).
- Chapter 6 details and outlines the first retrieval of cirrus cloud properties from upwelling radiance in the presence of cirrus using this retrieval method. These are the first retrievals of atmospheric and cloud properties performed using far-infrared radiance measurements for an aircraft platform, and have been submitted for publication in Panditharatne et al. (under review).
- Final conclusions for the work and further areas of investigation are discussed in Chapter 7.

Chapter 2

The Retrieval Method

The studies performed within this thesis are based on retrieval theory, which is an inverse method of extracting information from a measurement of a related quantity. This thesis uses the inverse method to extract temperature and water vapour profiles (Chapter 4 and 5) and ice cloud properties (Chapter 6) from upwelling radiances in the far- and mid-infrared by exploiting their spectral signals in these regions. This chapter is separated into two sections: Section 2.1 describes the radiative transfer theory that causes these spectral signals, and Section 2.2 details the inverse method used to perform the retrievals. A glossary of key symbols used within this chapter (Table A1 and A2) is contained within the Appendix.

2.1 Radiative Transfer

This section details the theory of radiative transfer and how the radiation emitted by the Earth is absorbed, re-emitted, and scattered as it passes through the atmosphere. This outline is based on the descriptions in Liou (2002), and a glossary of the symbols defined in this section can be found in Table A1 in the Appendix.

The change in intensity of the spectral dependent radiance dI as it passes a small distance, ds , through a medium such as the atmosphere is given by Lambert's law:

$$dI = -\sigma_e \rho I ds \quad (2.1)$$

where ρ is the density of the medium, and σ_e is the mass extinction cross section (in units of area per mass) and is a measure of how well the radiance is absorbed and scattered by the medium at a particular frequency. Both of these parameters can vary with distance through the medium.

Equation 2.1 shows that both scattering and absorption within the medium reduces the radiance as it passes through. However, if the medium is also emitting, it can increase the intensity of the radiation. Similarly, the multiple scattering of radiation from other locations into the direction of the beam will contribute to the intensity of dI . This introduces an additional source term into the right-hand side of Lambert's law, which can be defined using the source function coefficient, j , that has the same physical meaning as σ_e but for emission and multiple scattering. By adding this term to Eq. 2.1, it can be rearranged to

$$\frac{dI}{ds} = -\sigma_e \rho (I - J) \quad (2.2)$$

where $J \equiv j/\sigma_e$ and is the source function that is spectrally dependent.

This is the general form of the radiative transfer equation without any coordinate system.

When modelling atmospheres, it is useful to vertically divide the atmosphere into discrete layers using the plane-parallel approximation. This approximation makes three assumptions. The first is that the curvature of the Earth can be ignored. The second is that the medium is considered homogenous within each layer. The third is that I is isotropic. The linear distance normal to the layers can be denoted by z and is equivalent to the height or pressure axis of the atmosphere. Using this, the path of the radiation can be linked to the vertical axis using:

$$ds = \frac{dz}{\mu} \quad (2.3)$$

where $\mu = \cos(\theta)$, and θ is the zenith angle of the radiation's path.

It is convenient to define the optical depth, τ , as a measure of how opaque the atmosphere is between an altitude z and the top of the atmosphere, z_{TOA} , at a particular frequency:

$$\tau(z) = \int_z^{z_{\text{TOA}}} \sigma_e \rho(z) dz \quad (2.4)$$

where $\tau(z_{\text{TOA}}) = 0$.

This can be used to define the transmittance, $\mathcal{T}$, which is the fraction of the initial radiation that is transmitted over that path:

$$\mathcal{T}(\tau) = e^{-\tau}. \quad (2.5)$$

Using this new coordinate system, Eq. 2.2 can be rewritten as shown below.

$$\mu \frac{dI(\tau, \mu)}{d\tau} = I(\tau, \mu) - J(\tau, \mu) \quad (2.6)$$

To describe the upwelling radiation reaching a height z , the boundary conditions of τ at the surface and at a height of z are used to integrate Eq. 2.6 with the integrating factor ($e^{-\tau}$):

$$I^\uparrow(\tau(z), \mu) = I(\tau_{\text{surf}}, \mu) e^{-(\tau_{\text{surf}} - \tau(z))/\mu} + \int_{\tau(z)}^{\tau_{\text{surf}}} J(\tau', \mu) e^{-(\tau_{\text{surf}} - \tau(z))/\mu} \frac{d\tau'}{\mu} \quad (2.7)$$

where $0 < \mu \leq 1$ and τ_{surf} is the optical depth at the surface.

When Eq. 2.7 is used to model the Earth's OLR, the first term in Eq. 2.7 represents the extinction of the radiation emitted by the surface of the Earth. This can be represented using the spectral surface emissivity ϵ , and a Planck function $B(T)$ at the temperature of the surface (T_{surf}):

$$I^\uparrow(\tau(z), \mu) = \epsilon B(T_{\text{surf}}) e^{-(\tau_{\text{surf}} - \tau(z))/\mu} + \int_{\tau(z)}^{\tau_{\text{surf}}} J(\tau', \mu) e^{-(\tau_{\text{surf}} - \tau(z))/\mu} \frac{d\tau'}{\mu} \quad (2.8)$$

where $0 \leq \epsilon \leq 1$, and $\epsilon=1$ when the surface is emitting like a blackbody. The spectral surface emissivity and $B(T_{\text{surf}})$ are both also spectrally dependent.

A discussion of the theory related to the source function contributions will now be divided into two sections for clarity: the effects of atmospheric absorption and emission, and the scattering effects of ice clouds.

2.1.1 Atmospheric Absorption and Emission

Figure 1.2 shows the fine spectral features caused by the atmospheric gases absorbing and re-emitting the upwelling infrared radiation in clear-sky conditions. These fine structures are caused by energetic transitions associated with molecular vibration, translational, and rotational modes. If the energy of the incident radiation on a molecule matches that of a given energetic transition, then the incident radiation can be absorbed and re-emitted at a lower energy and so longer wavelength. This results in the spectral dependence of the absorption and emission of the radiation and reduces the transmittance of radiation through the atmosphere. Larger particles, such as aerosols, can also have a scattering effect on the outgoing infrared radiation, and this is considered in the next section.

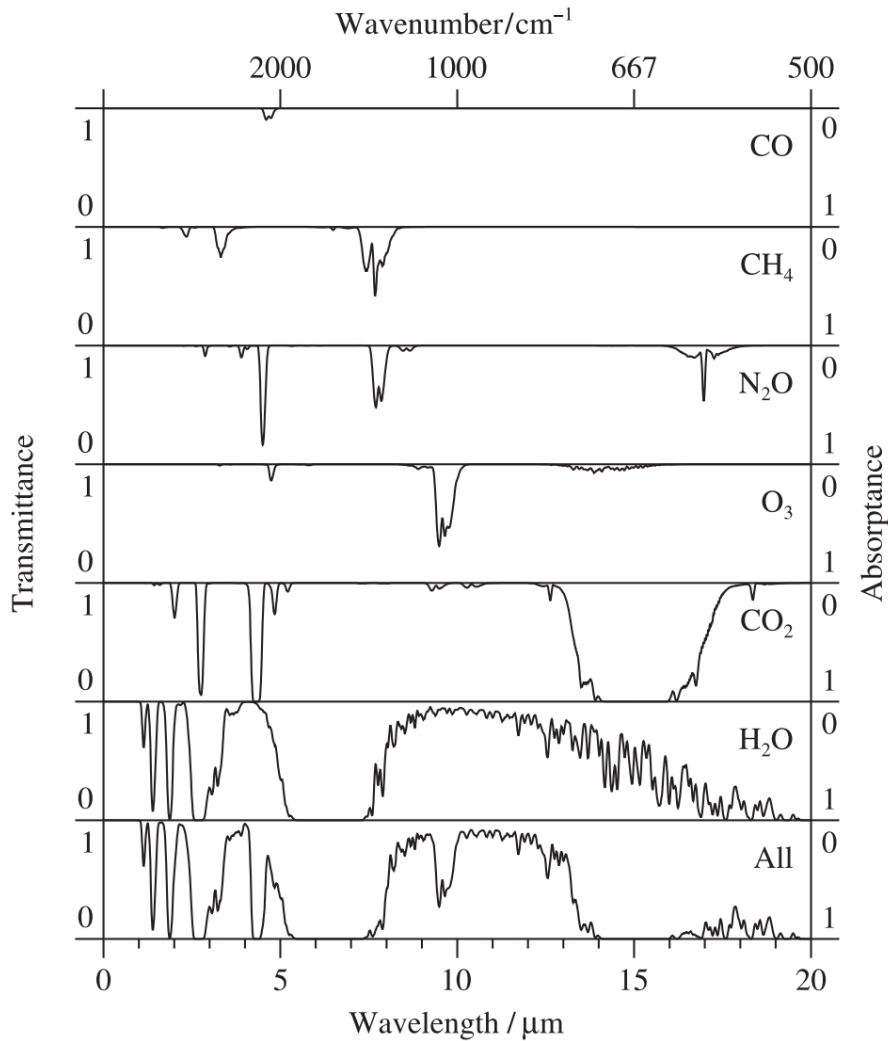


Figure 2.1: The transmittance and absorption of six strongly absorbing gases in the infrared, as well as for the six gases combined in the absence of clouds. Taken from Andrews (2010).

Figure 2.1 shows the spectrally dependent absorption of six gases present in the Earth's atmosphere that strongly absorb in the infrared. Between $6-667 \text{ cm}^{-1}$, the rotational band of water vapour dominates the absorption. CO_2 has a strong absorption band centred at 667 cm^{-1} , caused by the ν_2 symmetric vibrational mode. This is commonly referred to as the CO_2 $15 \mu\text{m}$ band, with the regions of decreasing absorption on either side referred to as the band wings. The region between $\sim 770-1250 \text{ cm}^{-1}$ is known as the atmospheric window, and is where the atmosphere is predominantly transparent, excluding the strong O_3 absorption at 1042 cm^{-1} and a limited absorption from CO_2 and water vapour. At higher wavenumbers, there are contributions from one of the water vapour vibration modes at 1595 cm^{-1} with a larger CO_2 absorption at $\sim 2325 \text{ cm}^{-1}$.

The atmospheric emission at each layer's temperature, attenuated by the optical thickness of that layer, contributes towards the upwelling radiance and so can be used to replace the source function in Eq. 2.8.

$$I^\uparrow(\tau(z), \mu) = \epsilon B(T_{\text{surf}}) e^{-(\tau_{\text{surf}} - \tau(z))/\mu} + \int_{\tau(z)}^{\tau_{\text{surf}}} B[T(\tau)] e^{-(\tau_{\text{surf}} - \tau(z))/\mu} \frac{d\tau'}{\mu} \quad (2.9)$$

This gives an equation that describes the upwelling infrared radiation observed at an altitude z that excludes the contributions of scattering.

2.1.2 The Scattering Effects of Ice Clouds

In order to consider the bulk radiative effect of an ice cloud, it is important to consider the properties of the ice crystals that compose it. The crystals within cirrus clouds range in size from $1 \mu\text{m}$ to 1 mm , and are distributed throughout the cloud through gravitational settling (Fu et al. 2017). The Ice Water Content (IWC) of the cloud represents the mass of ice particles per unit volume of air and varies from $0.0001-1.2 \text{ gm}^{-3}$ in ice clouds (Heymsfield et al. 2017). The IWC can also be integrated across the vertical extent of the cloud to get the Ice Water Path (IWP) which represents the mass of ice particles per unit area of air.

At the cloud top, ice crystals can take a variety of simple habits such as pristine hexagonal columns, plates, bullets and bullet-rosettes, shown in Figure 2.2 (Yang et al. 2005, Baran 2009). Larger ice crystals tend to form at warmer temperatures and sink lower within the cloud,

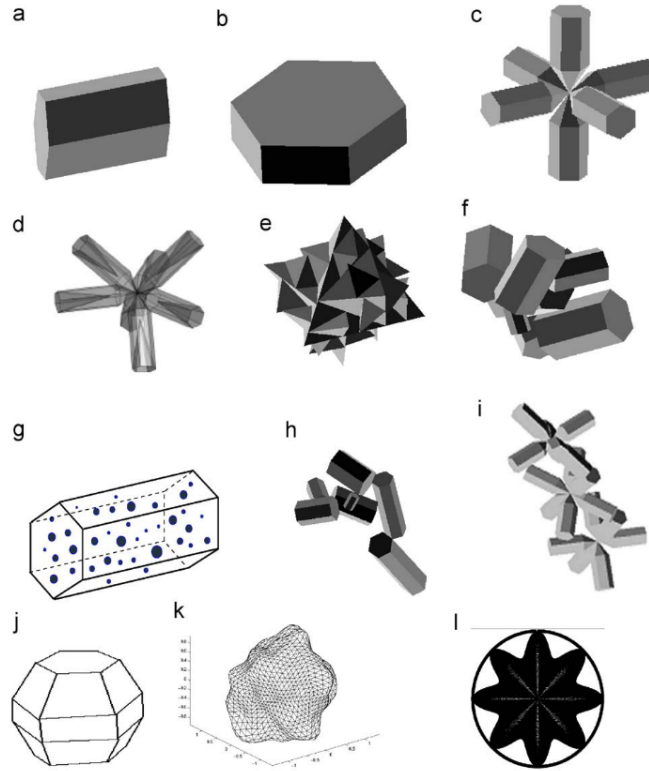


Figure 2.2: Ice crystal habits in ice clouds: (a) pristine hexagonal column, (b) pristine hexagonal plate, (c) six-branched bullet-rosette, (d) six-branched bullet-rosette with conical air cavities within each branch, (e) polycrystal, (f) hexagonal ice aggregate, (g) Inhomogeneous Hexagonal Monocrystal, (h) hexagonal chain aggregate, (i) rosette-aggregate, (j) droxtal, (k) Gaussian random sphere and (l) Chebyshev polynomial. Taken from Baran (2009).

growing with vapour deposition dependent on the amount of water vapour above or below the ice-saturated value (Heymsfield et al. 2017, Kärcher et al. 2014). At these lower altitudes in the cloud, ice crystals can also start to coalesce and form more complex aggregates of these habits (also shown in Figure 2.2) (Heymsfield 1975, van Diedenhoven & Kokhanovsky 2018). Current knowledge of the natural variation of ice crystal habits is still incomplete, and each habit can have varying scattering abilities (van Diedenhoven & Kokhanovsky 2018).

The refractive index of ice, shape, and size are the most significant factors in the single-scattering properties of an ice crystal (Zhang et al. 1999, Yang et al. 2005), and will be explored in the remainder of this subsection.

Single-Scattering Properties of Ice Crystals

For a spherical particle, the intensity and direction of scattering are dependent on its size and the wavelength of the incident radiation. The wavelengths associated with the OLR lie well

within the lower range of observed ice crystal sizes, and so the scattering is in the regime of Lorenz-Mie theory and geometric optics. In this regime, the incident radiation can be treated as a collection of separate parallel rays incident on a flat surface, and it is assumed that the spherical particle is composed of a homogeneous medium. These rays are able to undergo reflection, refraction, and diffraction around the particle, or can be absorbed through the surface depending on the medium's complex refractive index (Liou 2002).

The complex refractive index of ice is spectrally and temperature-dependent. The real component is related to the scattering of incident radiation, and the imaginary component (κ) is related to the absorption of incident radiation (Warren & Brandt 2008). Large values are indicative of high absorption and scattering, respectively, reducing transmission through an ice crystal, and can be used to calculate the mass absorption (σ_a) and scattering (σ_s) cross sections. Similar to σ_e , these are a measure of the radiation absorbed and scattered, respectively (in units of area per mass) by a medium at a particular frequency.

σ_a can be simply derived from the imaginary component of the refractive index:

$$\sigma_a = \frac{4\pi\kappa}{\lambda\rho} \quad (2.10)$$

where λ is the wavelength of the incident radiation.

σ_s is not only dependent on the size of the crystals and the real refractive index but also on the ice crystal habit. An exact solution for the scattering performed by these different ice crystal habits does not exist (Wendisch et al. 2007).

When radiation is scattered by a crystal, it scatters in all directions with an angular distribution of energies specific to the particle's orientation, shape, size, composition, and the incident wavelength (Liou 2002). This scattered radiation can contribute to intensities elsewhere, and the angular distribution of scattering is defined using the scattering phase function, $P(\mu)$:

$$\int_0^{2\pi} \int_0^\pi \frac{P(\mu)}{4\pi} \sin\theta d\theta d\phi = 1 \quad (2.11)$$

where ϕ is the azimuthal angle. The phase function can also be used to derive the

azimuthally averaged phase function, $\overline{P}(\mu, \mu_i)$:

$$\overline{P}(\mu, \mu_i) = \frac{1}{2\pi} \int_0^{2\pi} P(\mu, \phi, \mu_i, \phi_i) d\phi_i \quad (2.12)$$

where the subscript ‘i’ represents the incoming direction.

Computational methods such as the T-Matrix and Complex Angular Momentum (CAM) are used to calculate the optical properties of single crystals (Liou 2002). The T-matrix method considers the spherical electromagnetic waveforms of the incident and scattered fields using a matrix and is dependent on the real refractive index, size, shape, and orientation of the particle. It is particularly useful with non-spherical symmetrical ice crystals (Mishchenko et al. 1996). CAM theory uses the quantum mechanical tunnelling of photons to produce optical effects that would not be allowed with geometry optics or Mie theory (Nussenzveig & Wiscombe 1980).

Datasets have been developed from the above methods for remote sensing applications and reported in the solar, infrared, and microwave regions, with notable studies of the single-scattering properties in the entire infrared region including Baum et al. (2007) and Yang et al. (2005), with Baran & Francis (2004) examining aggregates, and Baran et al. (2001) examining hexagonal columns.

Bulk Cloud Properties

Three key bulk properties of ice clouds are considered in this work: the Cloud Effective Radius (CER), the Cloud Optical Thickness (COT), and the Cloud Top Height (CTH).

The impact of each crystal on the bulk optical properties of the cloud can be combined by integrating the single-scattering properties across the Particle Size Distribution (PSD) of crystals within the cloud for each crystal habit. The PSD describes the number concentration of ice crystals as a function of their size and is often defined at different altitudes through the cloud (Baran 2009, Baum et al. 2014, Yang et al. 2015). The CER is used as a measure of the PSD and is sometimes also referred to as the Cloud Effective Dimension. For the bulk cloud, it is a weighted mean of the size distribution of all the ice crystals:

$$\text{CER} = \frac{3}{2} \frac{V_{\text{tot}}}{A_{\text{tot}}} \quad (2.13)$$

where V_{tot} and A_{tot} are the total volume and total projected area, respectively, of the ice crystals within the bulk cloud (Baum et al. 2014). These are calculated for each type of ice crystal habit (h) using equations 2.14 and 2.15:

$$A_{\text{tot}} = \sum_{h=1}^M \left[\int_{D_{\min}}^{D_{\max}} A_h(D) f_h(D) n(D) dD \right] \quad (2.14)$$

and

$$V_{\text{tot}} = \sum_{h=1}^M \left[\int_{D_{\min}}^{D_{\max}} V_h(D) f_h(D) n(D) dD \right] \quad (2.15)$$

where M is the number of ice crystal habits observed in the cloud, $D_{\min}$ and $D_{\max}$ are the minimum and maximum sizes in the PSD, $A_h(D)$ and $V_h(D)$ are the projected area and volume of a specific crystal with habit and size (D), $f_h(D)$ is the fraction of ice crystals that have a given habit at each size, and $n(D)$ is the number distribution of sizes.

When averaged over the entire function, any two clouds with a given PSD can be composed of ice crystals with the same CER, but the shape of the distribution may differ. Varying proportions of small or large crystals flattens or sharpens the PSD in different regions. PSDs also tend to broaden with temperature and are particularly critical for cirrus clouds given their large range of crystal sizes (Heymsfield et al. 2017). The PSD impacts most of the microphysical and so radiative properties of clouds, and is vital in understanding its net radiative effect.

The PSD of a cloud can be used to define its bulk optical properties by defining the absorption (β_a), scattering (β_s), and extinction (β_e) coefficients of the cloud. These represent the spectrally dependent absorption, scattering, and extinction, respectively, of incident radiation travelling through a medium, for a unit length.

$$\begin{aligned}
 \beta_a &= \int \sigma_a(D) \rho n(D) dD \\
 \beta_s &= \int \sigma_s(D) \rho n(D) dD \\
 \beta_e &= \int \sigma_e(D) \rho n(D) dD
 \end{aligned}
 \tag{2.16}$$

These can be used to derive the bulk scattering albedo of the cloud, ω_0 :

$$\omega_0 = \frac{\beta_s}{\beta_e} = \frac{\beta_s}{\beta_a + \beta_s}
 \tag{2.17}$$

which represents the ratio of the scattering to extinction coefficients at a given wavenumber. It can also be referred to as the single scattering albedo of the cloud. When $\omega_0 = 1$, all incident radiation is scattered. When $\omega_0 = 0$, all incident radiation is absorbed.

The degree to which a cloud prevents the incident radiation at a given wavelength from passing through can then be defined using the COT:

$$\text{COT} = \int_{z_{\text{base}}}^{z_{\text{top}}} \beta_e dz
 \tag{2.18}$$

where z_{top} and z_{base} represent the altitudes at the top and base of the cloud, respectively.

A larger COT is indicative of a greater β_e and so a greater opacity at that wavenumber. Within the visible region, at $\lambda = 694 \text{ nm}$, cirrus with $\text{COT}_{694 \text{ nm}} < 0.03$ are referred to as sub-visible, $0.03 \leq \text{COT}_{694 \text{ nm}} \leq 0.3$ appear translucent and are known as thin cirrus, while $0.3 \leq \text{COT}_{694 \text{ nm}} \leq 3$ are opaque and appear white (Lynch et al. 2002).

One of the main reasons cirrus clouds are such a large uncertainty in climate models is because they can exhibit a range of optical thicknesses that are sensitive to their microphysics. For example, fewer but larger ice crystals reduce the COT, a property seen in clouds formed through heterogeneous nucleation (Kuebbeler et al. 2014, Lohmann et al. 2008), as does a faster sedimentation (Heymsfield et al. 2017). The COT is also partially dependent on κ , which for ice is high in regions of the OLR (Warren & Brandt 2008). This suggests a tendency

for a greater optical thickness in these regions, and so greater trapping of infrared radiation and a stronger greenhouse effect.

The net radiative effect of a cloud is also linked to its CTH. This refers to the highest visible part of the cloud from a nadir-viewing observer situated above the cloud. The CTH is indicative of the temperature at the top of the cloud and so directly relates to the wavelengths of radiation re-emitted by the cloud. It is also influential on the formation and microphysical properties of the cloud, as well as the sensitivity of the bulk radiative effect of the cloud to the microphysical properties. For example, Wendisch et al. (2007) highlighted that the radiative effect of a cirrus cloud with a low CTH but large COT is only slightly sensitive to ice crystal habit, while for high, optically thin cirrus, the infrared greenhouse effect at the TOA is much more dependent on ice crystal habit.

Using the scattering parameters derived in this section, the scattering effects of the cloud can now be included in the source function in Eq. 2.8 to give an equation for the upwelling radiances observed at an altitude z .

$$\begin{aligned}
 I^\uparrow(\tau(z), \mu) = & \epsilon B(T_{\text{surf}}) e^{-(\tau_{\text{surf}} - \tau(z))/\mu} && \text{Surface Emission} \\
 & + \int_{\tau(z)}^{\tau_{\text{surf}}} B[T(\tau)] e^{-(\tau_{\text{surf}} - \tau(z))/\mu} \frac{d\tau'}{\mu} && \text{Atmospheric Absorption and Emission} \\
 & + \frac{\omega_0}{2} \int_{-1}^1 I(\tau, \mu_i) \bar{P}(\mu, \mu_i) d\mu_i && \text{Scattering}
 \end{aligned} \tag{2.19}$$

2.1.3 Radiative Transfer Models

Radiative Transfer (RT) models are used to calculate the transfer of radiation through the atmosphere and, in this work, to simulate upwelling radiances. These work by solving equations 2.9 and 2.19 using a number of different methods and the user-input boundary conditions.

In this thesis, two types of RT models are used, both of which make certain assumptions to speed up the calculation process. The first type is a line-by-line RT model, which calculates the radiance at every spectral line for a given wavenumber range and resolution (Clough

et al. 2005). The second is a fast RT model which uses a parameterization of the line-by-line RT model outputs to increase computational speed (Saunders et al. 2018). This section will detail the line-by-line models used in this thesis, and the fast RT model will be detailed in Chapter 4.

LBLRTM

The Line-by-Line Radiative Transfer Model (LBLRTM) is used in this work to simulate infrared radiances in clear-sky conditions but does not include the scattering for cloudy scenes or interactions between shortwave radiation and atmospheric gases and aerosols (Clough et al. 2005). LBLRTM has been validated and improved using high-resolution spectroscopic measurements in the infrared (Mlawer et al. 2019), and is considered the gold standard for radiative transfer codes (Kratz et al. 2005, Oreopoulos et al. 2012).

LBLRTM can be used to perform calculations at different observing altitudes and angles. The temperature and water vapour profiles, as well as up to 38 other optional gas species, are defined on user-input, or default, pressure or altitude levels. The boundary conditions such as temperature, emissivity, and reflectivity can also be specified if necessary. Using these, LBLRTM can be used to output spectrally resolved radiances and transmittance from the desired viewing altitudes, as well as the spectrally dependent optical depth of each layer (Clough et al. 2005).

The absorption effects of each gaseous species are based on the spectroscopic line parameters from High-resolution Transmission Molecular Absorption (HITRAN) (Gordon et al. 2022). The absorption of radiation by molecules away from the centre of discrete spectral lines, known as continuum absorption, is defined using the MT_CKD model. This includes the continuum effects of water vapour (self- and foreign broadening), ozone, carbon dioxide, nitrogen, and oxygen (Mlawer et al. 2019).

LBLRTM+DISORT (LBLDIS)

To simulate upwelling radiances that include the effects of scattering, the clear-sky outputs from the LBLRTM code are combined with the DIScrete-Ordinate Radiative Transfer Model

(DISORT) code. The combined LBLRTM and DISORT models will be referred to as the LBLRTM+DISORT (LBLDIS) model (Stamnes et al. 2000, Turner & Holz 2005). Currently, DISORT is considered one of the most accurate radiative transfer algorithms to treat the multiple scattering effect, and has been used to evaluate new radiative transfer codes in the infrared (DiNatale et al. 2020, Martinazzo et al. 2021).

The spectrally dependent layer optical depths output from LBLRTM are input into DISORT, alongside the user input temperature and boundary conditions. The CER and number density or bulk extinction coefficient at a defined wavenumber are inserted into DISORT onto the same vertical levels defined in LBLRTM, and it is assumed that each layer is homogeneous. These are then used to select the scattering properties from pre-defined databases.

DISORT then uses the discrete ordinate method to transform Eq. 2.19 into a set of simultaneous equations that can be solved. To do this, it discretizes the directional variation of the radiation using the phase function into streams. Solutions are then found for each stream, and combined to encompass the total solid angle range of 4π (Stamnes et al. 2000).

2.2 Optimal Estimation Retrievals

This section details the inverse method used within this thesis to extract information about the vertical temperature and water vapour profile and the properties of ice clouds. A detailed description of various methods for solving these problems can be found in Rodgers (2000), however this work only uses the optimal estimation method. The following section presents a summary based on Rodgers (2000), and outlines the fundamental theory and terminology that is used within this thesis.

The general inverse problem can be separated into two spaces: measurement and state space, as shown in Figure 2.3. In this case, the measurement space contains the upwelling radiances within the measurement vector, $\mathbf{y}$. In this thesis, state space contains information about the atmospheric and surface conditions. Within it, the true state vector, $\mathbf{x}$, contains information about the retrieval targets, and is composed of a combination of vertical temperature and water vapour profiles, surface skin temperature, surface emissivity, and cloud

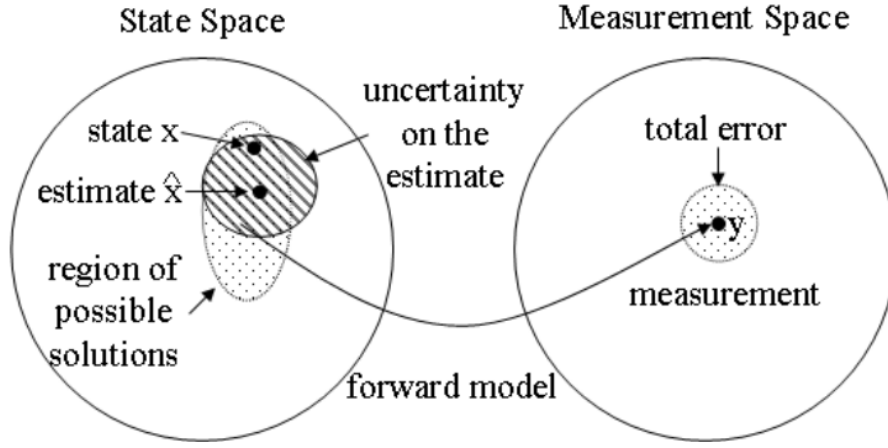


Figure 2.3: The link between state and measurement space is used to solve the general inverse problem. The forward model translates the measurement vector (y) into state space. The associated measurement error is propagated into a region of possible solutions. The true state x should lie within this region, and the estimated $\hat{x}$ must capture it within its uncertainties. Taken from Rodgers (2000).

properties. These vectors are linked through a forward function, $f(x)$, that can describe the complete physics of the measurement, and calculate y from x as in Eq. 2.20:

$$y = f(x, b) + \epsilon \quad (2.20)$$

where ϵ is the experimental error in the measured radiance, and b represents known quantities that influence the measurement such as other gases like ozone or carbon dioxide.

The forward function itself is approximated using a forward model, $F(x, b)$:

$$F(x, b) = f(x, b, b') + \Delta f \quad (2.21)$$

where b' represents the forward function parameters that have been excluded or approximated in the forward model, and Δf is the error in the forward model relative to the real physics. In this work, the forward model is a RT model that can simulate upwelling radiances (measurement space) given certain assumptions stored in b and information about the atmosphere and surface (state space).

As shown in Figure 2.3, the uncertainty that is introduced into the system from both the forward model and instrument uncertainty gives rise to a range of possible solutions

from the retrieval method, as multiple states in the state space can map to multiple points in measurement space. For example, if the radiance at a given wavenumber increases with temperature but is inversely proportional to water vapour concentrations, then there are different combinations of the two variables that can result in the measured radiance.

Bayes theorem can be used to solve this inverse problem by reducing the number of possible solutions, producing a probability density function (PDF) of the state given a particular measurement:

$$P(\mathbf{x}|\mathbf{y}) = \frac{P(\mathbf{y}|\mathbf{x})P(\mathbf{x})}{P(\mathbf{y})} \quad (2.22)$$

where:

- $P(\mathbf{x}|\mathbf{y})$ is the probability density function of the state given a particular measurement and is the target of the retrieval.
- $P(\mathbf{y}|\mathbf{x})$ represents the information about $\mathbf{y}$ which would be obtained by a measurement of $\mathbf{x}$ and can be calculated using the forward model and knowledge of the measurement error.
- $P(\mathbf{x})$ is the a-priori knowledge of the state and indicates that the quantity $P(\mathbf{x})d\mathbf{x}$ is the probability before the measurement that $\mathbf{x}$ is in the interval $(\mathbf{x}, \mathbf{x}+d\mathbf{x})$.
- $P(\mathbf{y})$ is in practice a normalising factor that does not need to be calculated, but similarly indicates that the quantity $P(\mathbf{y})d\mathbf{y}$ is the probability before the measurement that $\mathbf{y}$ is in the interval $(\mathbf{y}, \mathbf{y}+d\mathbf{y})$.

The relationship between $\mathbf{x}$ and $\mathbf{y}$ is non-linear, however it is convenient to first linearise Eq. 2.20 around the first-guess state, $\mathbf{x}_0$, using a first-order Taylor expansion, and substitute in Eq. 2.21.

$$\mathbf{y} - \mathbf{F}(\mathbf{x}_0) = (\mathbf{x} - \mathbf{x}_0) \left. \frac{\partial \mathbf{F}}{\partial \mathbf{x}} \right|_{\mathbf{x}=\mathbf{x}_0} = \mathbf{K}_x(\mathbf{x} - \mathbf{x}_0) + \epsilon \quad (2.23)$$

where $\mathbf{K}_x = \partial \mathbf{F}(\mathbf{x}, \mathbf{b}) / \partial \mathbf{x}$ is a weighting function matrix that contains the derivatives of the forward model outputs (radiances) with respect to each retrieval product. It, therefore,

represents the sensitivity of the forward model to the state.

For a linear case, the measurement statistics are Gaussian, and so $P(\mathbf{y}|\mathbf{x})$ can be expressed as:

$$-2\ln P(\mathbf{y}|\mathbf{x}) = (\mathbf{y} - \mathbf{K}_x \mathbf{x})^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{K}_x \mathbf{x}) + c_1 \quad (2.24)$$

where c_1 is a constant, $\mathbf{S}_y$ is the measurement covariance matrix, the superscript ' T ' indicates the transpose, and the superscript ' -1 ' indicates the matrix inverse. The measurement covariance represents a combination of the uncertainty that arises from the observation and the forward model's calculation. The diagonal elements of the measurement covariance represent the uncorrelated uncertainty between each element of $\mathbf{y}$, and the off-diagonals represent the correlation between elements. For measured spectra, this can be derived from the instrument uncertainty and through comparisons to the forward model.

$P(\mathbf{x})$ can be written as:

$$-2\ln P(\mathbf{x}) = (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a) + c_2 \quad (2.25)$$

where c_2 is a constant, $\mathbf{x}_a$ is the a-priori value of $\mathbf{x}$, and $\mathbf{S}_a$ is the a-priori covariance matrix that represents the potential variability in the state.

By substituting equations 2.24 and 2.25 into Eq. 2.22, the PDF of the atmospheric state for a given measurement can be obtained:

$$-2\ln P(\mathbf{x}|\mathbf{y}) = (\mathbf{y} - \mathbf{K}_x \mathbf{x})^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{K}_x \mathbf{x}) + (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a) + c_3. \quad (2.26)$$

This is also a Gaussian PDF with a covariance, $\mathbf{S}_x$, given by:

$$\mathbf{S}_x = (\mathbf{S}_a^{-1} + \mathbf{K}_x^T \mathbf{S}_y^{-1} \mathbf{K}_x)^{-1} \quad (2.27)$$

where the square-root-diagonal of this matrix represents the estimated standard deviation (ESD) or retrieval error associated with the best estimate of the state, $\hat{\mathbf{x}}$, that is calculated by

minimising the right-hand side of Eq. 2.26 giving:

$$\begin{aligned}\hat{\mathbf{x}} &= \mathbf{x}_a + (\mathbf{K}_x^T \mathbf{S}_y^{-1} \mathbf{K}_x + \mathbf{S}_a^{-1})^{-1} \mathbf{K}_x^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{K}_x \mathbf{x}_a) \\ &= \mathbf{x}_a + \mathbf{G} (\mathbf{y} - \mathbf{K}_x \mathbf{x}_a)\end{aligned}\tag{2.28}$$

where $\mathbf{G}$ is the gain matrix that shows the influence of the measurement on the retrieval.

Non-linearity, as in this case, increases the complexity of the forward model, and so the first-order derivative of the forward model outputs ($\mathbf{K}_x$) cannot be used. Assuming that the measurement and a-priori errors remain Gaussian, the best estimate of the state must now be found iteratively by minimising the right-hand side of Eq. 2.26. This can be done by evaluating the right-hand side of Eq. 2.26 into a cost function, $\mathbf{J}$, which has a χ^2 distribution:

$$\mathbf{J}(\mathbf{x}) = (\mathbf{y} - \mathbf{F}(\mathbf{x}, \mathbf{b}))^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{F}(\mathbf{x}, \mathbf{b})) + (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a) + c_3 \tag{2.29}$$

where c_3 is constant.

This cost function can be split into two parts corresponding to the first two expressions on the right-hand side of Eq. 2.29. The first term is the measurement cost, $\mathbf{J}_y$, and represents the fit of the forward model output to the measurement vector within the limits of $\mathbf{S}_y$. A high measurement cost indicates a poorer fit and suggests that the retrieved state vector is less likely to match the true state. The state cost, $\mathbf{J}_x$, indicates how far the retrieved state has deviated from the a-priori in relation to $\mathbf{S}_x$. A high state cost could be caused by either a poor a-priori, which is not similar to the true state, or a small a-priori covariance, which suggests the retrieval is tightly constrained to the a-priori estimate.

Minimising $\mathbf{J}$ requires the use of numerical methods and a-priori knowledge of the state to constrain the first estimate. The optimal estimation method iterates from the a-priori state through estimates to minimise Eq. 2.29 and reach the final solution. Generally, two different approaches are used to minimise $\mathbf{J}$ and they are contained within the Levenberg-Marquardt method (Marquardt 1963):

$$\begin{aligned} \mathbf{x}_{i+1} &= \mathbf{x}_i - [\Delta_{\mathbf{x}}^2 \mathbf{J}(\mathbf{x}_i) - \gamma_i \mathbf{I}_n]^{-1} \Delta_{\mathbf{x}} \mathbf{J}(\mathbf{x}_i) \\ &= \mathbf{x}_i + (\mathbf{K}_{\mathbf{x},i}^T \mathbf{S}_y^{-1} \mathbf{K}_{\mathbf{x},i} + \mathbf{S}_a^{-1} + \gamma_i \mathbf{I}_n)^{-1} [\mathbf{K}_{\mathbf{x},i}^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{F}(\mathbf{x}_i, \mathbf{b})) + \mathbf{S}_a^{-1} (\mathbf{x}_a - \mathbf{x}_i)] \end{aligned} \quad (2.30)$$

where the subscript ‘ i ’ represents each iteration number, $\mathbf{x}_{i+1}$ represents each iteration of the state vector, $\Delta_{\mathbf{x}}$ represents the derivative with respect to $\mathbf{x}_{i+1}$, γ_i is the Levenberg-Marquardt parameter used for each iteration, $\mathbf{I}_n$ is the $n \times n$ identity matrix, and $\mathbf{K}_{\mathbf{x},i}$ represents each iteration of the weighting function matrix.

The initial Levenberg-Marquardt parameter can be found experimentally for different retrievals. However, generally at the beginning of the retrieval, γ is large, and Eq. 2.30 turns into the method of steepest descent:

$$\begin{aligned} \mathbf{x}_{i+1} &= \mathbf{x}_i - \frac{1}{\gamma} \Delta_{\mathbf{x}} \mathbf{J}(\mathbf{x}_i) \\ &= \mathbf{x}_i + (\gamma_i \mathbf{I}_n)^{-1} [\mathbf{K}_{\mathbf{x},i}^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{F}(\mathbf{x}_i, \mathbf{b})) + \mathbf{S}_a^{-1} (\mathbf{x}_a - \mathbf{x}_i)] \end{aligned} \quad (2.31)$$

While the criteria for the adjustment of γ can vary, generally with each step, if the new value of $\mathbf{J}$ has decreased, the step is accepted, and γ is decreased. If the minimum has been overstepped, and $\mathbf{J}$ increases, then γ increases and the estimate $\mathbf{x}_i$ is left unchanged.

As $\gamma \rightarrow 0$, Eq. 2.30 becomes:

$$\begin{aligned} \mathbf{x}_{i+1} &= \mathbf{x}_i - [\Delta_{\mathbf{x}} \mathbf{J}(\mathbf{x}_i)]^{-1} \mathbf{J}(\mathbf{x}_i) \\ &= \mathbf{x}_i + (\mathbf{K}_{\mathbf{x},i}^T \mathbf{S}_y^{-1} \mathbf{K}_{\mathbf{x},i} + \mathbf{S}_a^{-1})^{-1} [\mathbf{K}_{\mathbf{x},i}^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{F}(\mathbf{x}_i, \mathbf{b})) + \mathbf{S}_a^{-1} (\mathbf{x}_a - \mathbf{x}_i)] \end{aligned} \quad (2.32)$$

where successive approximation steps are used to iterate through estimates of the zero value and local gradients using the Newton-Raphson method.

The optimal estimation solution will then occur once $\mathbf{J}$ has reached a minimum, or it has converged based on a selected criteria. Optimal estimation models may also stop if a maximum number of iterations have been reached. If $\mathbf{J}$ has not converged, the retrieval did not work and more information is required.

2.2.1 Characterising the retrievals

To evaluate the quality of the retrieval framework and retrieved products, two comparisons can be performed: a comparison of the retrieved state to the a-priori (Section 2.2.2), and a comparison of the retrieved state to the true state (Section 2.2.3) if the information is available. This can be done by considering a function $\mathbf{R}$ that represents the retrieval method based on the non-linear equivalent of Eq. 2.28:

$$\hat{\mathbf{x}} = \mathbf{R}(\mathbf{y}, \hat{\mathbf{b}}, \mathbf{x}_a, \mathbf{c}) \quad (2.33)$$

where $\hat{\mathbf{b}}$ represents the best estimate of the forward function parameters, and $\mathbf{c}$ contains other parameters that are not in the forward function but do affect the retrieval. It is important to note that in a well-performing retrieval method, it is assumed that $\partial \mathbf{R} / \partial \mathbf{c} = 0$ as these excluded parameters should not have a significant impact on the retrieval.

The retrieved value can now be related to the true state by substituting Eq. 2.20 and 2.21 into Eq. 2.33.

$$\hat{\mathbf{x}} = \mathbf{R}(\mathbf{F}(\mathbf{x}, \mathbf{b}) + \Delta f(\mathbf{x}, \mathbf{b}, \mathbf{b}') + \epsilon, \hat{\mathbf{b}}, \mathbf{x}_a, \mathbf{c}) \quad (2.34)$$

This is a configuration of a transfer function that describes the operation of the observing system, involving both the instrument and the retrieval method. This enables the characterisation of the sensitivity of the retrieval to the other state vectors and each source of error.

2.2.2 A comparison of the retrieved state to the a-priori state

The first step in exploiting the transfer function is substituting a linearisation of the forward model about $\mathbf{x} = \mathbf{x}_a$, $\mathbf{b} = \hat{\mathbf{b}}$ into Eq. 2.34:

$$\hat{\mathbf{x}} = \mathbf{R}(\mathbf{F}(\mathbf{x}_a, \hat{\mathbf{b}}) + \mathbf{K}_x(\mathbf{x} - \mathbf{x}_a) + \mathbf{K}_b(\mathbf{b} - \hat{\mathbf{b}}) + \Delta f(\mathbf{x}, \mathbf{b}, \mathbf{b}') + \epsilon, \hat{\mathbf{b}}, \mathbf{x}_a, \mathbf{c}) \quad (2.35)$$

where $\mathbf{K}_x$ represents the sensitivity of the forward model as discussed, and $\mathbf{K}_b = \partial \mathbf{F}(\mathbf{x}, \mathbf{b}) / \partial \mathbf{b}$

and represents the sensitivity of the forward model to the parameters.

Next, a linearisation of the forward model can be applied with respect to y , and $y' = F(x_a, b)$.

$$\begin{aligned}\hat{x} &\simeq R|_{y=y'} + \frac{\partial R}{\partial y}|_{y=y'}(y - y') \\ &= R(F(x_a, b), \hat{b}, x_a, c) + G[K_x(x - x_a) + K_b(b - \hat{b}) + \Delta f(x, b, b') + \epsilon]\end{aligned}\quad (2.36)$$

G is the same gain matrix as in Eq. 2.28 and represents the sensitivity of the retrieval to the measurement and its error where $G = \partial R / \partial y$.

By subtracting x_a from Eq. 2.36 the retrieved value can be compared to the a-priori state:

$$\begin{aligned}\hat{x} - x_a &= R(F(x_a, b), \hat{b}, x_a, c) - x_a && \text{Bias} \\ &+ A(x - x_a) && \text{Smoothing} \\ &+ G\epsilon_y && \text{Measurement error}\end{aligned}\quad (2.37)$$

where A is the sensitivity of the retrieved to the true state:

$$A = \frac{\partial \hat{x}}{\partial x} = GK_x \quad (2.38)$$

and ϵ_y is the total error associated with the forward model and instrument:

$$\epsilon_y = K_b(b - \hat{b})\Delta f(x, b, b') + \epsilon \quad (2.39)$$

The three terms on the right-hand side of Eq. 2.37 can be broken down to evaluate the error in the retrieval method associated with a bias in the method, smoothing as a result of the retrieval method's resolution, and the measurement error.

The first term on the right-hand side of Eq. 2.37 is the bias associated with a retrieval from a measurement simulated using the a-priori state and the forward model. A non-zero value

suggests that the retrieval method is not functioning and that there is an inherent bias in the method. Therefore, a zero value is expected as a requirement, where:

$$\mathbf{R}(\mathbf{F}(\mathbf{x}_a, \hat{\mathbf{b}}), \hat{\mathbf{b}}, \mathbf{x}_a, \mathbf{c}) = \mathbf{x}_a. \quad (2.40)$$

and must also be true when $\mathbf{x}_a$, $\hat{\mathbf{b}}$, or $\mathbf{c}$ are perturbed.

Given this, Eq. 2.37 can be rearranged to express the impact of the smoothing and measurement error on the retrieved state.

$$\begin{aligned} \hat{\mathbf{x}} &= \mathbf{x}_a + \mathbf{A}(\mathbf{x} - \mathbf{x}_a) + \mathbf{G}\epsilon_y \\ &= (\mathbf{I}_n - \mathbf{A})\mathbf{x}_a + \mathbf{A}\mathbf{x} + \mathbf{G}\epsilon_y. \end{aligned} \quad (2.41)$$

In the ideal case, $\mathbf{A}$ would be a unit matrix, and the retrieved state would be a combination of the true state and measurement error term, and there would be no impact of smoothing. In reality, the rows of $\mathbf{A}$ can be regarded as smoothing functions or averaging kernels (AK) that in this case peak at specific vertical levels with a half-width that is a measure of the resolution of the retrieval. The columns of $\mathbf{A}$ give the response of the retrieval to a δ function perturbation to the state vector and can be used to calculate the averaging kernel matrix. The area of each averaging kernel is approximately unity at levels where the retrieval is accurate, and a larger averaging kernel represents a greater amount of retrieval that comes from the measurement rather than the a-priori. Similarly a small averaging kernel suggests the retrieval has relied more on the a-priori.

The impact of the measurement error on the retrieved state will be explored further through a comparison of the retrieved state to the true state.

2.2.3 A comparison of the retrieved state to the true state

By rearranging equations 2.37 and 2.39, an expression for the difference between $\hat{\mathbf{x}}$ and $\mathbf{x}$ can be found and separated into different sources of error in the retrieval process.

$$\begin{aligned}
 \hat{\mathbf{x}} - \mathbf{x} &= (\mathbf{A} - \mathbf{I}_n)(\mathbf{x} - \mathbf{x}_a) && \text{Smoothing} \\
 &+ \mathbf{G}\mathbf{K}_b(\mathbf{b} - \hat{\mathbf{b}}) && \text{Model Parameter} \\
 &+ \mathbf{G}\Delta\mathbf{F}(\mathbf{x}, \mathbf{b}, \mathbf{b}') && \text{Forward Model} \\
 &+ \mathbf{G}\epsilon && \text{Retrieval Noise}
 \end{aligned} \tag{2.42}$$

Typically in retrievals, the true state is an unknown and so the mean of an ensemble of states is used, and a covariance matrix describes the statistics of the error. In this work, the true state is either known or there is a good approximation of it. Therefore, the smoothing error shown by the first term on the right-hand side of Eq. 2.42 can be used to calculate the best profile that could be retrieved within the constraints of the vertical resolution and a-priori of the retrieval framework. This best profile will be referred to as $\mathbf{x}_{AK}$, where

$$\mathbf{x}_{AK} = \mathbf{x}_a + \mathbf{A}(\mathbf{x} - \mathbf{x}_a) = \mathbf{x} + (\mathbf{A} - \mathbf{I}_n)(\mathbf{x} - \mathbf{x}_a). \tag{2.43}$$

In the ideal case, $\mathbf{x}_{AK} = \mathbf{x}$, and no smoothing from the retrieval framework will have occurred.

If smoothing the true state significantly reduces the residual between the true and retrieved state i.e $|\hat{\mathbf{x}} - \mathbf{x}_{AK}| \ll |\hat{\mathbf{x}} - \mathbf{x}|$ then the difference between the retrieved and true state is likely linked to the a-priori and vertical resolution. If this is not the case, and the residuals are comparable, this suggests the differences could be related more to the measurement.

The second term on the right-hand side of Eq. 2.42 represents the model parameter error. If the parameters in the forward model have been estimated correctly, and the model is linear for each of the individual parameters, then the expected error will be zero. If this is not the case then the error covariance, $\mathbf{S}_f$, is:

$$\mathbf{S}_f = \mathbf{G}\mathbf{K}_b\mathbf{S}_b\mathbf{K}_b^T\mathbf{G}^T \tag{2.44}$$

where $\mathbf{S}_b$ is the error covariance matrix for $\mathbf{b}$ and is determined separately for both

random and systematic errors.

The third term on the right-hand side of Eq. 2.42 represents the forward model error. This is associated with the approximations used within the model. In this work, it is approximated using test simulations in comparison to slower but more accurate RT models, LBLRTM and LBLDIS.

The final term on the right-hand side of Eq. 2.42 represents the retrieval noise. This is calculated by using the measurement noise and is normally unbiased and uncorrelated as a result.

$$\mathbf{S}_m = \mathbf{G}\mathbf{S}_y\mathbf{G}^T \quad (2.45)$$

where $\mathbf{S}_m$, is the covariance of the retrieval noise.

2.2.4 Additional Retrieval Diagnostic Parameters

One of the major advantages of the optimal estimation method is that it outputs other diagnostic parameters of the retrieval alongside the cost function and the retrieval error that were introduced in Section 2.2. These diagnostic parameters can be used to assess the amount of information the retrieval has about its target, and be used to optimise the retrieval. This is particularly useful within this work to identify the advantage of including the far-infrared. The following examples are two common diagnostic outputs for optimal estimation that are used in this thesis.

The Degrees of Freedom, $\mathbf{D}_F$, is a scalar measure of the number of pieces of information in a set of measurements that are independent of the a-priori. Using Eq. 2.29, this can be divided into two parts:

$$\mathbf{D}_{F,n} = \xi\{(\mathbf{y} - \mathbf{F}(\mathbf{x}, \mathbf{b}))^T \mathbf{S}_y^{-1}(\mathbf{y} - \mathbf{F}(\mathbf{x}, \mathbf{b}))\}; \mathbf{D}_{F,s} = \xi\{(\hat{\mathbf{x}} - \mathbf{x}_a)^T \mathbf{S}_a^{-1}(\hat{\mathbf{x}} - \mathbf{x}_a)\} \quad (2.46)$$

where ξ is the expected value operator, $\mathbf{D}_{F,s}$ is the degrees of freedom of the signal (DOFS), and $\mathbf{D}_{F,n}$ is the degrees of freedom of the noise (DOFN).

$\mathbf{D}_{F,s}$ can be explicitly calculated by using the relation for the trace of a product of two matrices $tr(\mathbf{CD}) = tr(\mathbf{DC})$ and considering Eq. 2.27. In practice, it is calculated using the gain matrix.

$$\begin{aligned}\mathbf{D}_{F,s} &= \xi \{tr[(\hat{\mathbf{x}} - \mathbf{x}_a)^T (\hat{\mathbf{x}} - \mathbf{x}_a) \mathbf{S}_a^{-1}]\} \\ &= tr(\mathbf{S}_{\hat{\mathbf{x}}} \mathbf{S}_a^{-1}) \\ &= tr([\mathbf{K}^T \mathbf{S}_y^{-1} \mathbf{K} + \mathbf{S}_a^{-1}]^{-1} \mathbf{K}^T \mathbf{S}_y^{-1} \mathbf{K}) = tr(\mathbf{GK})\end{aligned}\tag{2.47}$$

The same can be applied to $\mathbf{D}_{F,n}$:

$$\mathbf{D}_{F,n} = [\mathbf{S}_y(\mathbf{F}(\mathbf{x}, \mathbf{b}) \mathbf{S}_a \mathbf{F}(\mathbf{x}, \mathbf{b})^T + \mathbf{S}_y)^{-1}]^T\tag{2.48}$$

A low DOFS indicates that there is too little information in the measurement about one of the state vector elements or that two or more of the state vector elements are highly correlated. Therefore, the larger DOFS the better.

The Information Content of a Measurement, $\mathbf{H}$, is the factor by which knowledge of a quantity is improved by making a measurement.

$$\mathbf{H} = -\frac{1}{2} \ln |\mathbf{I} - \mathbf{A}|\tag{2.49}$$

The measurement will approach maximum information content as $\mathbf{A}$ approaches an identity matrix.

The theory and parameters introduced in Section 2.2 are used within this thesis to perform optimal estimation retrievals on far- and mid-infrared radiances to extract information about water vapour and ice clouds. These observed radiances are measured using Fourier transform spectrometers, and so the theory behind these instruments and observations will be described in the next chapter.

Chapter 3

Building FORUM-aircraft Observations

The aircraft-level observations of upwelling radiances used within this thesis were taken using two Fourier transform spectrometers. This chapter will explore the theory (Section 3.1) and instruments (Section 3.2) used to measure these radiances, as well as how they have been adapted to mimic the spectral characteristics of FSI observations (Section 3.3).

3.1 Fourier Transform Spectroscopy

The fundamentals of Fourier transform spectroscopy will be explored in this section through a simple two-beam Michelson interferometer, shown in Figure 3.1. This can, in general, be applied to other designs of Fourier transform spectrometers and is taken from textbooks written by Griffiths & de Haseth (1986) and Davis et al. (2001).

The setup for a simple two-beam Michelson interferometer is shown in Figure 3.1. Incident radiation on the aperture of the interferometer is collimated before passing through a beam splitter, which divides the beam into two. Each of these beams is reflected by respective mirrors. While one mirror is fixed, the other can be moved, introducing a variable optical path difference (OPD) between the two beams of light. The reflected beams are then recombined by the same beam splitter. This recombined beam is incident on the detector that measures the final intensity of the recombined beam. When the OPD is an integer multiple of the original beam's wavelength, the two beams are coherent, and the intensity of the combined beam will be at a maximum. When the OPD is a half-integer multiple of the wavelength, the two beams

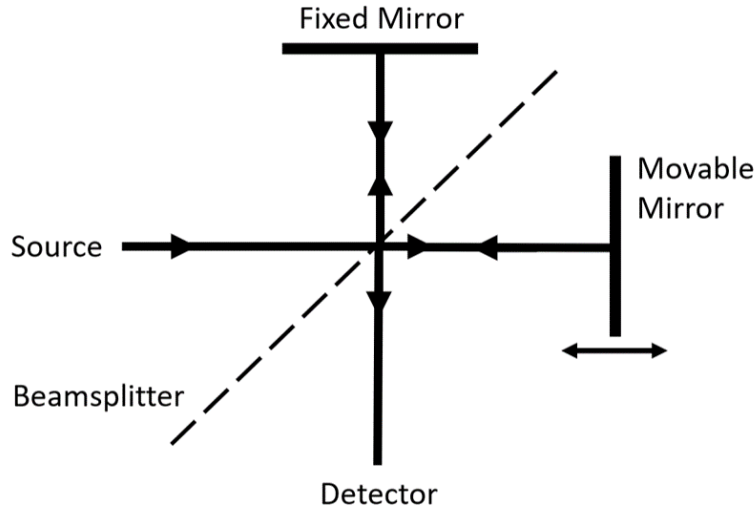


Figure 3.1: A schematic of a two-beam Michelson interferometer.

are incoherent, and the intensity of the combined beam will be at a minimum. If the mirror is scanned continuously, the signal that reaches the detector varies sinusoidally as a function of OPD. This signal is called the interferogram and is the output of an interferometer.

By considering just the varying part of the signal, and removing any DC offset, a mathematical expression for the interferogram, I_δ , can be derived for a monochromatic source:

$$I_\delta = I_\nu \cos(2\pi\nu\delta) \quad (3.1)$$

where δ is the OPD, ν is the wavenumber, and I_ν is the measured radiance. It is important to note that I_ν is affected by the instrument characteristics such as the beamsplitter material and optical alignment.

For a broadband source of radiation, Eq. 3.1 can be integrated across all the frequencies contained within the broadband source:

$$I_\delta = \int_{-\infty}^{\infty} I_\nu \cos(2\pi\nu\delta) d\nu. \quad (3.2)$$

The measured spectrum can then be found by performing a Fourier transform on Eq. 3.2:

$$I_\nu = \int_{-\infty}^{\infty} I_\delta \cos(2\pi\nu\delta) d\delta. \quad (3.3)$$

An interferometer with an infinite OPD would observe an infinitely long interferogram

and produce a spectrum with an infinitely high resolution. This would result in idealised spectral absorption features that appear in the measured spectrum as infinitely narrow spectral lines. However, this is not possible in practice. For a finite OPD that is assumed to be symmetrical, the maximum and minimum values can be defined as $\pm\Delta$ (with units of length), and results in the truncation of the interferogram using a truncation function, $D(\delta)$, commonly known as a boxcar function:

$$\begin{aligned} D(\delta) &= 1 & \text{if } -\Delta \leq \delta \leq \Delta, \\ D(\delta) &= 0 & \text{if } \delta > |\Delta|. \end{aligned} \tag{3.4}$$

Assuming that the measured interferogram is symmetrical, the Fourier transform of the truncated interferogram is then found to be:

$$I_\nu = \int_{-\infty}^{\infty} I_\delta D(\delta) \cos(2\pi\nu\delta) d\delta \tag{3.5}$$

This truncation of the interferogram results in the convolution of the idealised spectrum with a function that is a Fourier transform of $D(\delta)$:

$$f(\nu) = 2\Delta \operatorname{sinc}(2\pi\nu\delta). \tag{3.6}$$

This is a sinc function, $f(\nu)$, with a height of 2Δ and a Full Width Half Maximum (FWHM) of $0.605/\Delta$. This is sometimes sufficient to represent an interferometer's instrument line shape (ILS), which represents the instrument's response to an infinitely narrow spectral line.

This process results in a large 'ringing' effect in the spectrum around the spectral lines that are narrow compared to the ILS. Additional processing can be done to remove this 'ringing' through a process called apodisation, where the corresponding sinc function is convolved with an apodisation function to get a best fit of the observed ILS that compromises between resolution and reduced 'ringing'. Any function that has a value of 1 at $\delta = 0$ and decreases to zero as $|\delta|$ increases can be used as an apodisation function.

In many cases, the true OPD is not known exactly due to a finite sampling interval or a frequency dependence of the OPD that is introduced by instrument components. This leads

to an uncertainty in the point of zero phase difference, resulting in the measurement of an asymmetric interferogram. To correct for this, a complex Fourier transform can be applied to the measured interferogram to gain the complex form of the measured spectrum, I'_ν :

$$I'_\nu = \int_{-\infty}^{\infty} I_\delta e^{-i2\pi\nu\delta} d\delta. \quad (3.7)$$

The phase between the real and imaginary components of I'_ν is defined by the phase angle, θ_ν , which typically slowly varies with frequency and can be calculated using:

$$\theta_\nu = \arctan \left(\frac{\text{Im}(I'_\nu)}{\text{Re}(I'_\nu)} \right) \quad (3.8)$$

where $\text{Re}(I'_\nu)$ and $\text{Im}(I'_\nu)$ represent the real and imaginary components of the spectrum, respectively.

If the phase angle is zero, all of the spectral information is present in the real part of the spectrum, and no correction is required. If the phase angle is non-zero, some of the spectral information is dispersed into the imaginary plane. To retrieve this information, a phase correction can be applied using the following equation:

$$I_\nu = \text{Re}(I'_\nu)\cos(\theta_\nu) + \text{Im}(I'_\nu)\sin(\theta_\nu). \quad (3.9)$$

As the phase angle only slowly varies with wavenumber, it is often calculated at a low spectral resolution and then interpolated to the resolution of the full spectrum for the phase correction.

Finally, it is important to consider that any spectrum measured by an interferometer is affected by its instrument response function, R_ν . The true spectrum, L_ν , can be calculated using:

$$I_\nu = R_\nu L_\nu + C_\nu \quad (3.10)$$

where C_ν is an offset that can be caused by the instrument self-emission or atmospheric emission. If C_ν is assumed constant for the duration of the radiance measurements, then

the instrument response function can be calculated by finding the difference between two measurements of known radiance. This is usually performed using two calibration targets that are held at a hot and ambient temperature. If the calibration targets are assumed to have a uniform temperature and a spectral emissivity of $\epsilon_{\nu, BB}$, the instrument response function can be derived using Eq. 3.11:

$$R_{\nu} = \frac{I_{\nu, HBB} - I_{\nu, ABB}}{\epsilon_{\nu, BB} B_{\nu}(T_{HBB}) - \epsilon_{\nu, BB} B_{\nu}(T_{ABB})} \quad (3.11)$$

where $I_{\nu, HBB}$ and $I_{\nu, ABB}$ are the measured radiances from the hot and ambient calibration targets, respectively, and $B_{\nu}(T_{HBB})$ and $B_{\nu}(T_{ABB})$ are the expected blackbody radiances associated with the temperatures of the hot and ambient calibration targets, respectively.

The measurements of the radiances emitted by these calibration targets, commonly referred to as the calibration views, are typically interspersed between measurements of the scene. The closest calibration views in time to the scene measurements are then used to calculate the instrument response function.

The true radiances of the scene can then be derived from the measured radiances by combining Eq. 3.10 and 3.11:

$$L_{\nu} = \frac{I_{\nu} - I_{HBB}}{R_{\nu}} + B(T_{HBB}). \quad (3.12)$$

3.2 Instruments Used in This Work

The observational data that will be analysed within this thesis were taken during two science flights. The first science flight analysed in this work is the C153 flight (13 March 2019) which is explored in Chapter 5. The second science flight is the B895 flight (13 March 2015), which is explored in Chapter 6. The radiance observations taken during these flights were measured by the following instruments and are available on the Centre for Environmental Data Analysis (CEDA) Archive.

Tropospheric Airborne Fourier Transform Spectrometer (TAFTS)

TAFTS is a four-port Martin-Puplett interferometer (Canas et al. 1997). A Martin-Puplett interferometer uses the same setup as the Michelson interferometer but uses wire grid polarizers instead of mirrors to flip the polarization of the light beam. Measurements are made at the output ports by pairs of detectors, each containing a "longwave" and "shortwave" detector made of a Germanium-Gallium alloy (GeGa) and silicon antimonide (SiSb), respectively. The instrument has two pairs of blackbody calibration targets held at ambient temperature and 323 K. There is an internal calibration before scans, with a single nadir scan taking ~ 1.5 s with an angular field of view of 1.6 deg. TAFTS has a spectral range of nominally 80-300 cm^{-1} (longwave channel) and 330-600 cm^{-1} (shortwave channel) with a sampling of 0.06 cm^{-1} . It has a nominal spectral resolution of 0.12 cm^{-1} that can be reduced to 0.24 cm^{-1} , by reducing the optical path difference and scan time, to reduce the instrument noise and allow more spectra to be collected.

The calibrated TAFTS data were provided by Jonathan Murray (Imperial College London) with the accompanying random noise and calibration uncertainties. The raw interferograms were first processed using an IDL routine developed by Paul Green and are described in Beeby (2011). This routine first interpolates the raw interferograms onto the same spatial sampling scale to account for variations in the movement of the TAFTS scanning mirror. The centre of each interferogram was then identified, and a complex Fourier transform was applied to it to produce complex spectra and derive the phase angle (Section 3.1). The final step of the routine was to 'de-ghost' the spectra to remove the effects of vibrations of the beamsplitter by applying a filter function at higher wavenumbers. The spectra were then calibrated and phase-corrected using a method developed by Jon Murray that is described in Murray (2019). This method isolated the self-emission of the instrument by calculating the differences between the measured radiances of the calibration target pairs.

One of the TAFTS spectrum that is used in Chapter 5 is shown in blue in Figure 3.2a. The TAFTS spectrum that is used in Chapter 6 is shown in blue in Figure 3.3. The uncertainty on a single TAFTS spectrum is composed of the random noise and calibration error added in quadrature. The random noise is a combination of the photon noise, caused by the inherent

random timing of photons being received and counted by the detector, and the random vibrations the instrument experiences by being stationed on an aircraft. There are also additional peaks in the NESR at 360 and 440 cm^{-1} due to absorption from the beamsplitter, and at 460 cm^{-1} due to the polypropylene view window. The random noise for the C153 and B895 flights is shown in Figure 3.2b and Figure 3.3b, respectively. The calibration error was estimated using the estimated self-emission of the instrument and the uncertainty in the temperatures of the blackbody calibration targets. The TAFTS calibration error for both the C153 and B895 flights is shown in Figure 3.2c and Figure 3.3c, respectively. In the shortwave channel, the final measurement uncertainty during both flights is of the order of $\pm 1\text{-}2 \text{ mW m}^{-2} \text{ sr}^{-1} / \text{cm}^{-1}$ and in the longwave channel is substantially less than this. In both channels, the uncertainty increases towards the detector band edges.

Airborne Research Interferometer Evaluation System (ARIES)

ARIES uses a Michelson-type configuration with a mercury cadmium telluride (HgCdTe) photodetector for the "longwave" channel and an indium antimonide (InSb) photodetector for the "shortwave" channel (Wilson et al. 1999). It also has two temperature-controlled blackbody targets and performs periodic calibrations during its measurement sequence. A single ARIES scan takes $\sim 0.25 \text{ s}$ and has an angular field of view of 2.5° . The instrument spectral range covers 550-1800 cm^{-1} (longwave channel) and 1700-3000 cm^{-1} (shortwave channel), with a spectral resolution of 1 cm^{-1} and a spectral sampling of 0.42 cm^{-1} . Only observations from the longwave channel are considered here, as the shortwave channel exceeds the FSI spectral range.

The calibrated ARIES data were provided by Stuart Fox (Met Office) with the accompanying random noise and calibration uncertainties. A single ARIES scan from the C153 flight that is used in Chapter 5 is shown in green in Figure 3.2a. A single ARIES scan that is used in Chapter 6 is shown in green in Figure 3.3a. The ARIES measurement uncertainty also consists of the random noise and calibration error. For the C153 flight, the ARIES noise (Figure 3.2b) and calibration (Figure 3.2c) uncertainties were provided in radiance units. For the B895 flight, the random noise (Figure 3.3b) and calibration (Figure 3.3c) uncertainties were approx-

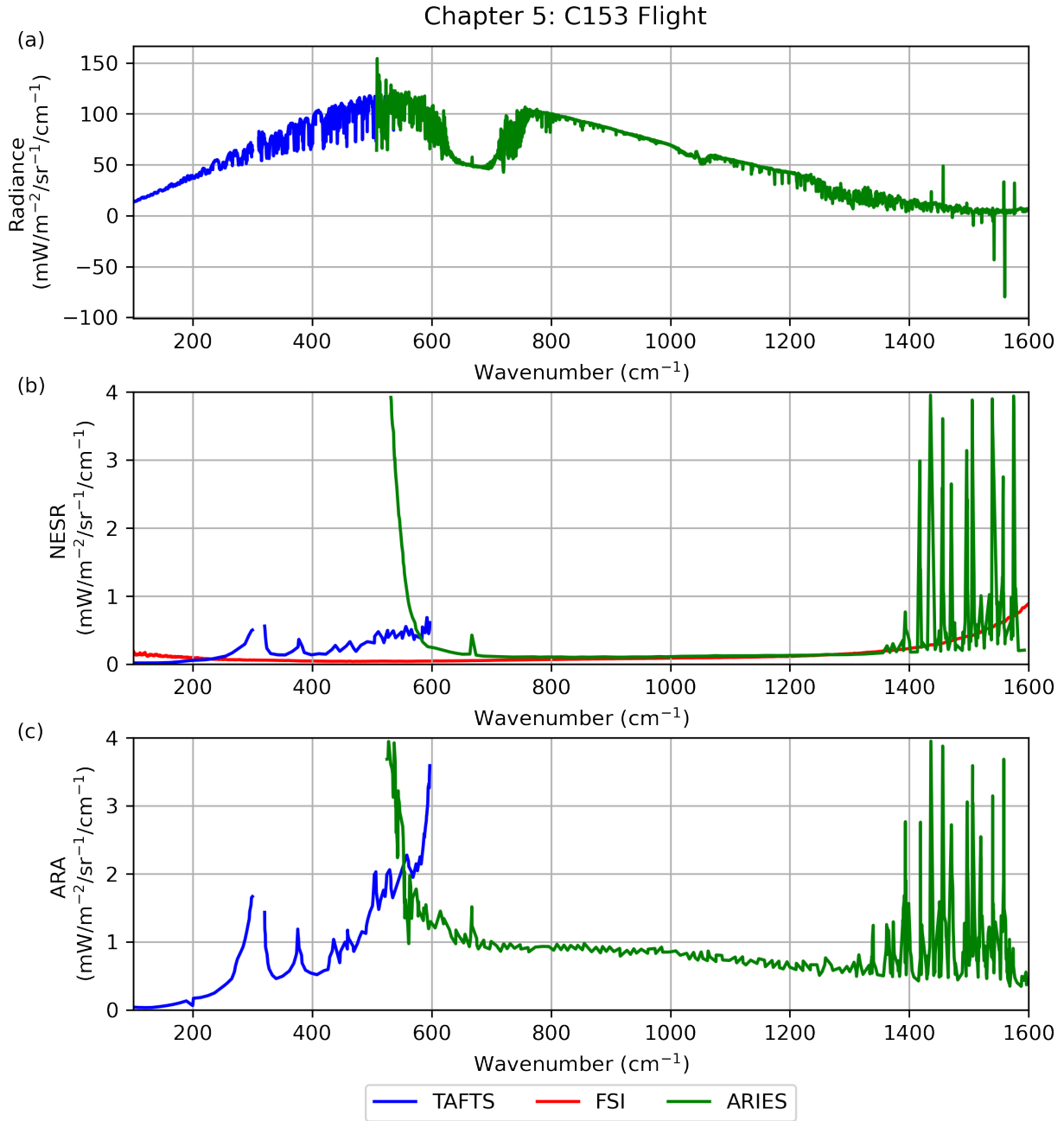


Figure 3.2: A summary of the uncertainties related to radiance observations taken by TAFTS and ARIES that are used in Chapter 5. (a) An example of a clear-sky spectrum (single scan) observed by TAFTS and ARIES during the C153 flight. (b) The Noise-Equivalent-Spectral-Radiance (NESR) and (c) Absolute Radiometric Accuracy (ARA) for TAFTS and ARIES from Warwick et al. (2022), as well as the expected NESR values for the FSI. The target ARA for the FSI described is in brightness temperature, and is 0.1 K between 300-1100 cm^{-1} , 0.2 K between 200-300 cm^{-1} and 1100-1300 cm^{-1} , and 1 K elsewhere.

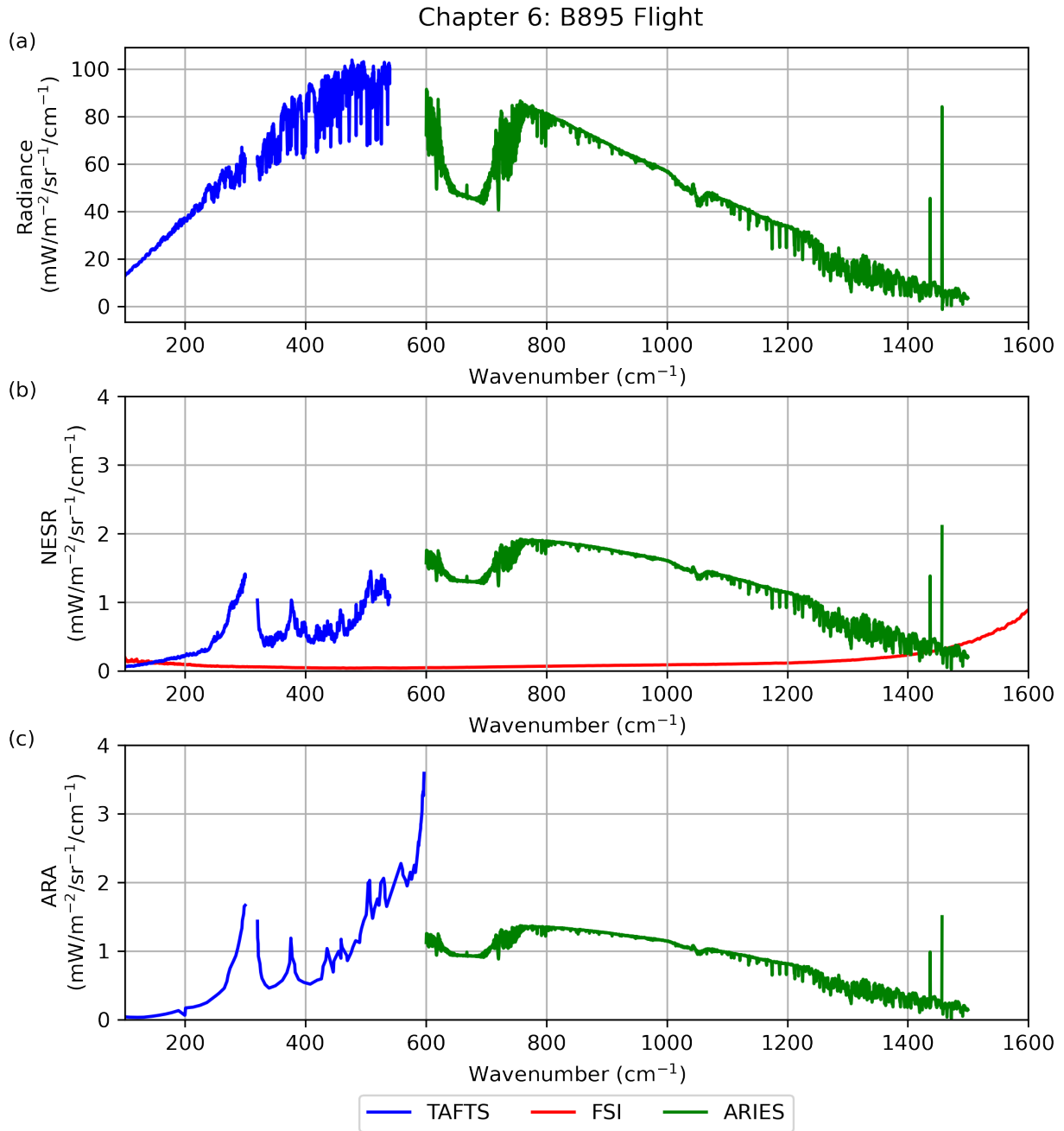


Figure 3.3: A summary of the uncertainties related to radiance observations taken by TAFTS and ARIES that are used in Chapter 6. (a) An example of a cloudy spectrum (single scan) observed by TAFTS and ARIES during the B895 flight. (b) The Noise-Equivalent-Spectral-Radiance (NESR) and (c) Absolute Radiometric Accuracy (ARA) for TAFTS and ARIES from Bantges et al. (2020), as well as the expected NESR values for the FSI. The target ARA for the FSI described is in brightness temperature, and is 0.1 K between 300-1100 cm^{-1} , 0.2 K between 200-300 cm^{-1} and 1100-1300 cm^{-1} , and 1 K elsewhere.

imations provided through personal communication to Richard Bantges (Imperial College London) (Bantges et al. 2020). These were given in brightness temperature (BT) and were assumed constant in brightness temperature throughout the ARIES longwave channel. The random noise was approximated to be 1.4 K for a single scan, and the calibration uncertainty was approximated to be 1 K. These have been converted to radiance units based on the spectrum shown in Figure 3.3a.

Between $650\text{-}1350\text{ cm}^{-1}$, the final measurement uncertainty (random noise and calibration uncertainty combined in quadrature) for a single ARIES scan can reach up to $\pm 2.5\text{ mW m}^{-2}\text{ sr}^{-1}/\text{cm}^{-1}$ for both the C153 and B895 flights. Large spikes in the ARIES NESR and ARA are also observed above 1300 cm^{-1} , reaching up to $\pm 4\text{ mW m}^{-2}\text{ sr}^{-1}/\text{cm}^{-1}$ during the C153 flight. These spikes were caused by a strong water vapour absorption that was occurring as a result of excess water vapour in the optical path of the instrument. This excess water vapour was caused by outgassing of the materials used to build the instrument and so occurred during both campaigns (Bantges et al. 2020).

3.3 Constructing FORUM-aircraft Observations

Combined, TAFTS and ARIES cover the full FSI spectral range, and there are a number of aircraft campaigns that have included coincident observations taken by both of these instruments. This presents a unique opportunity to assess the potential of what spectral information could be stored within the FSI observations and provide an idea of what the FORUM mission could deliver. Therefore, the TAFTS and ARIES observations have been altered to mimic the FSI ILS and sampling to create ‘FORUM-aircraft’ observations.

To do this, the ILS of TAFTS, ARIES, and the FSI must first be considered, and the best approximations of them are shown in Figure 3.4. TAFTS, ARIES, and the FSI are all Fourier transform spectrometers and so output interferograms that undergo a Fourier transform to acquire the measured spectra following the theory outlined in Section 3.1. ARIES has an OPD of 1.01 cm, and so a top-hat function of width 1.01 cm is applied to the interferogram. In wavenumber space, this translates to the Fourier transform of a top-hat function - a sinc

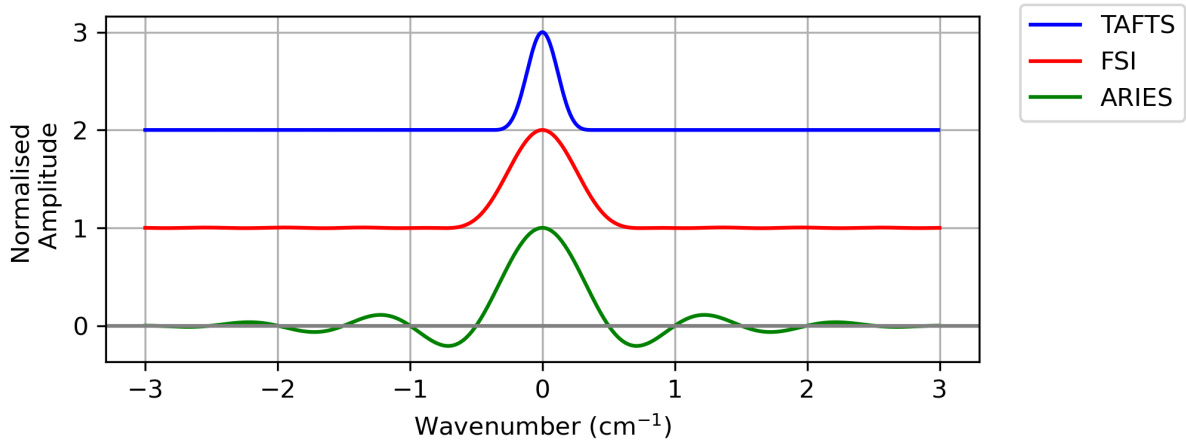


Figure 3.4: Instrument line shapes for (green) ARIES, (blue) TAFTS, and (red) FSI that are used in this work. The instrument line shapes are offset for TAFTS (+2) and the FSI (+1) for easier comparison.

function. This results in a large ‘ringing’ effect in the spectrum around the peak, shown in green in Figure 3.4. For ARIES, this is seen in the observed spectra, and so, as a first iteration, this can be taken as the ILS for ARIES. For TAFTS, this ringing is not observed in its measured spectra due to the high resolution of the instrument and the pressure broadening of the atmospheric absorption lines beneath the aircraft. For the FSI, this ringing is also not currently expected based on the target instrument resolution. Therefore, for TAFTS and the FSI, an apodisation function is used to better model their respective ILS. In this work, TAFTS uses an apodisation function composed of a Kaiser and top-hat function as this was iteratively found to produce the best fit to the real ILS, and the FSI uses the strong Norton-Beer apodisation function based on the expected instrument requirements (Norton & Beer 1976), resulting in the ILSs shown in Figure 3.4.

The FORUM-aircraft observations have been constructed by applying the FSI ILS to the observed TAFTS and ARIES radiances using the process highlighted in purple in Figure 3.5. ARIES does not have an apodisation function in this work, therefore, the FSI apodisation function can be directly applied to the ARIES spectra. TAFTS, on the other hand, must have its apodisation function removed before the FSI apodisation function can be applied. The spectral characteristics of both instruments will likely introduce an error into the final FORUM-aircraft observations that needs to be investigated to properly constrain the retrieval.

To assess this error, high-resolution simulated spectra have been treated in two ways

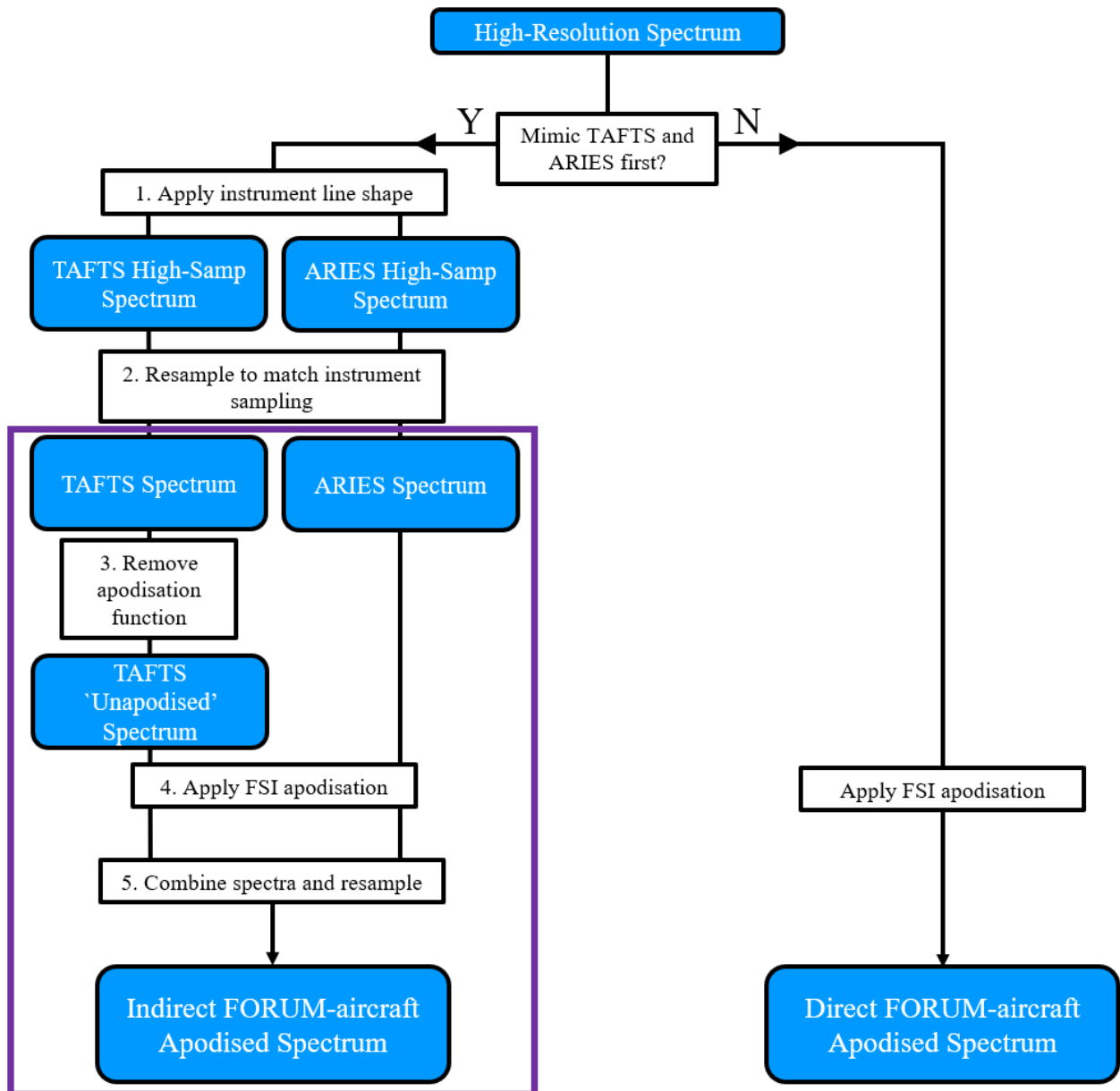


Figure 3.5: The process used to develop FORUM-aircraft observations or simulations. The whole flowchart is used to calculate the error associated with the apodisation process. The path on the right-hand side describes the 'direct' process of applying the FSI apodisation to a high-resolution simulated spectrum, where radiances aren't modified to look like TAFTS and ARIES first. The path on the left-hand side describes the 'indirect' process where the TAFTS and ARIES spectral characteristics are applied, the TAFTS spectral characteristics are removed, and then the FSI apodisation is applied. The steps applied to the TAFTS and ARIES observations are outlined in purple.

outlined through the two main pathways in Figure 3.5. First, following the right-hand pathway, the FSI ILS has been directly applied. Second, following the left-hand pathway, the individual TAFTS and ARIES ILS have been applied to the simulation and have been resampled onto their corresponding sampling grids. The apodisation function for TAFTS has

then been removed, and the FSI characteristics have then been applied to both the TAFTS and ARIES simulations. By taking the difference between these two methods (left-hand path and right-hand path in Figure 3.5, the impact of the original instrument characteristics on the FORUM-aircraft measurements can be evaluated. This work examines both clear and cloudy spectra, and so the process has been tested on an LBLRTMv12.11 simulation of the TAFTS and ARIES spectrum examined in Warwick et al. (2022), which relates to the work in Chapter 5, and on an LBLDIS simulation of a TAFTS and ARIES spectrum from Bantges et al. (2020), which relates to the work in Chapter 6. The residuals are shown in Figure 3.6 in radiance units alongside the instrument uncertainties from the B895 flight from Figure 3.3. It is important to recall that for ARIES, the noise and calibration uncertainty were assumed constant in brightness temperature throughout the longwave channel.

The impact of the FORUM-apodisation on the ARIES simulations are considered first. On a fine sampling scale, the apodisation process on the ARIES simulation causes residuals that mostly remain within the instrument uncertainty for a clear-sky case (green in Figure 3.6b). In reality, ARIES observations have a coarser sampling of 0.42 cm^{-1} , and as Figure 3.6b highlights, this significantly increases the associated residuals. This can be explained by considering the loss of information associated with a coarser sampling of spectral features; the peak of a curve is lost if it is only sampled along its increasing edges. This effect is most prominent in the CO_2 wings, which are typically exploited in retrievals to obtain the vertical temperature profile. This effect is slightly reduced for the cloudy case. However, this larger uncertainty is likely to degrade the performance of the temperature retrieval, with potential knock-on effects for simultaneous retrievals of water vapour and cloud properties.

TAFTS, in contrast, has a fine sampling of 0.06 cm^{-1} and a finer resolution of 0.24 cm^{-1} , and so does not experience the same issues as ARIES. However, it has two other areas of concern. First, its own apodisation function must be removed before the FSI apodisation can be applied (Step 3 in Figure 3.5). The residuals between the ‘unapodised’ TAFTS spectrum and the original high-resolution simulation were less than $1 \text{ mW m}^{-2} \text{ sr}^{-1} / \text{cm}^{-1}$ for both clear and cloudy cases. Figure 3.4 highlights that as the FSI ILS is much broader than the TAFTS ILS, this lost spectral information would be removed by the FSI apodisation and so

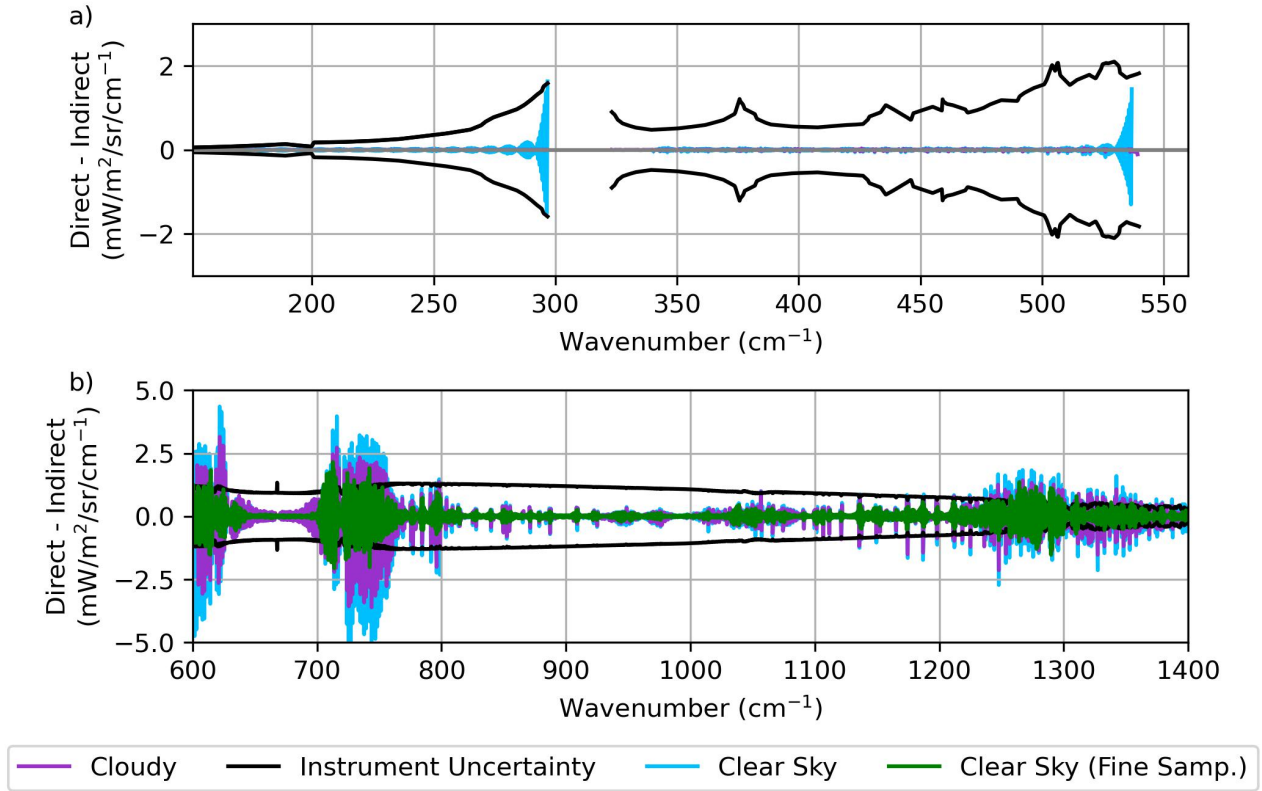


Figure 3.6: The uncertainty associated with developing FORUM-aircraft observations in radiance units. The light blue line (clear-sky) and purple line (cloudy) show the residuals between LBLRTMv12.11 or LBLDIS simulations of the TAFTS and ARIES observations that have had the FSI apodisation directly applied and been made to look like (a) TAFTS or (b) ARIES first. These simulations are based on observations from Warwick et al. (2022) and Bantges et al. (2020). The green line shows the clear-sky residual where the simulated ARIES spectrum has a finer sampling (Fine Samp.) than in the real case before the FSI apodisation has been applied. The black line is the noise and calibration uncertainty combined in quadrature associated with the ARIES longwave channel and both TAFTS channels from Figure 3.3b and c for the B895 flight.

does not affect the FORUM-aircraft spectrum.

Second, both of the TAFTS channels are used within this work. In the direct approach, the FSI apodisation is applied across the entire FSI spectral range, distributing the sampling of the points of the apodisation function across the entire target spectral range. The TAFTS observations are provided separately for the longwave and shortwave channels, which both cover a shorter spectral range than the FSI. This shorter spectral range means that the sampled points of the apodisation function are distributed differently across the spectra between the indirect and direct approaches. These differences result in the observed pattern in the BT residuals between the direct and indirect approaches in Figure 3.6a. This method is required,

given the separate TAFTS channels. However, this effect is not significant as the values remain within the original measurement uncertainty.

Following this process, coincident TAFTS and ARIES spectra can be converted into FORUM-aircraft observations. These will match the expected spectral characteristics of the FSI instrument; however, they are limited by the field of view, scan time, and corresponding instrument uncertainties. This uncertainty is also increased by the apodisation process and will be considered in the measurement covariance used to constrain the retrieval.

Chapter 4

Extending the Infrared Microwave Sounding Retrieval Scheme into the Far-Infrared

Key Points

- This chapter details the extension of the Infrared Microwave Sounding retrieval scheme for use on the FORUM Sounding Instrument.
- New coefficients and covariance matrices have been introduced into the retrieval scheme based on the instrument requirements.
- A database of 240 clear-sky profiles has been built using MERRA-2 reanalysis data to provide a testing set for changes to the retrieval scheme.
- The outputs of clear-sky retrievals from IASI and FSI simulated spectra showed that an additional DOFS was observed for both temperature and water vapour when using the FSI configuration, however this is in part due to the idealised measurement covariance of the FSI.

4.1 Introduction

This chapter outlines the extension of the RAL Infrared Microwave Sounding (IMS) optimal estimation retrieval scheme into the far-infrared for use on upcoming FSI observations. Section 4.2 describes the retrieval scheme that was developed by Richard Siddans (RAL Space), which, prior to this thesis, was not able to perform retrievals in the far-infrared. Section 4.3 outlines the full structure of the extension performed by the author of this thesis. Section 4.4 describes the 240 test cases used to monitor iterations made to the retrieval framework. Section 4.5 assesses the current ability of the retrieval framework, and technical changes are made in Section 4.6. The optimisation of the framework is then described in Section 4.7, with conclusions in Section 4.8.

4.2 Infrared Microwave Sounding (IMS)

The Infrared Microwave Sounding retrieval scheme developed at RAL Space uses the optimal estimation method described in Section 2.2 to simultaneously retrieve vertical profiles of atmospheric temperature and gases, along with surface skin temperature, surface spectral emissivity, and cloud parameters (Siddans 2019). It was initially developed for joint retrievals of observations from IASI, as well as the Microwave Humidity Sounder (MHS), and the Advanced Microwave Sounding Unit (AMSU) on the MetOp satellites, and is a customisable retrieval framework that can be adapted to a variety of instruments. This section will describe the retrieval framework and default settings that it currently uses.

A brief overview of the process used within IMS is summarised in Figure 4.1, however full details of the IMS algorithm can be found in Siddans (2019) and Trent et al. (2023). Richard Siddans (RAL Space) performed the work described in this subsection to configure IMS for use on IASI observations.

IMS uses the optimal estimation method from Rodgers (2000) (described in Section 2.2) to fit an observed spectrum (the measurement vector, $\mathbf{y}$) by iteratively perturbing the retrieval targets ($\mathbf{x}$). Estimations of observed spectra are calculated from adjusted values of $\mathbf{x}$ using a forward model, $\mathbf{F}(\mathbf{x})$, which in this case is a RT model called Radiative Transfer for the TIROS

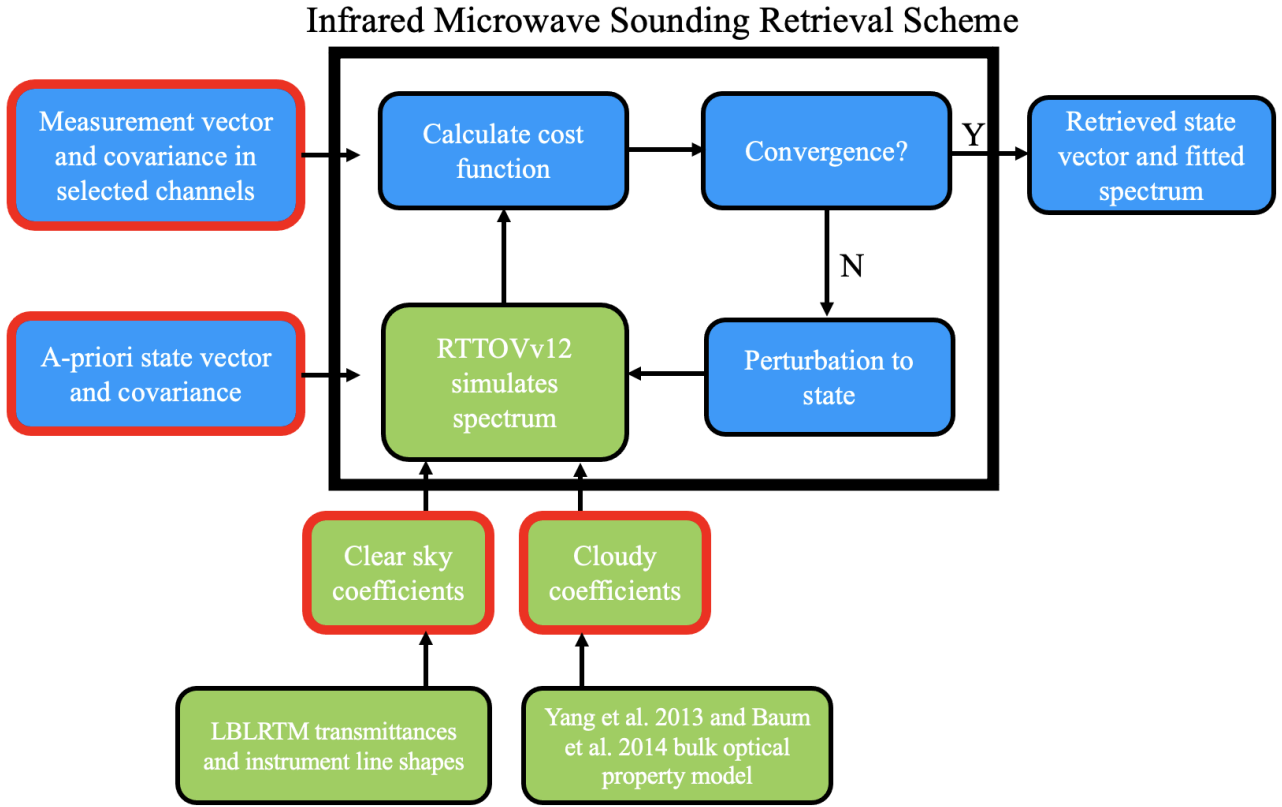


Figure 4.1: A schematic summarising the optimal estimation process within IMS. The observed spectrum and a-priori to the state are input into IMS. Using the radiative transfer model (RTTOVv12) the retrieval framework simulates a first guess spectrum. Components of RTTOVv12 are highlighted in green and described in Section 4.2.2. This spectrum is compared against the measured spectra at certain wavenumbers (channels), and a cost function is calculated. IMS works to minimise this cost function by adjusting the state to ultimately output the retrieved state and fitted spectrum. Components highlighted in red are spectrally dependent.

Operational Vertical Sounder v12 (RTTOVv12) (Saunders et al. 2018). Prior knowledge of the state is contained in the a-priori state vector, $\mathbf{x}_a$, with its covariance, $\mathbf{S}_a$, representing the variability and correlation of the profile, and both are used to constrain the retrieval. Similarly, the measurement covariance, $\mathbf{S}_y$, represents the uncertainty in the measurement and, for IASI, has been calculated based on the residual between simulations and bias-corrected IASI observations in the current IMS configuration. IMS uses an eigenvector representation of the state vector to increase computational speed during the retrieval process (Trent et al. 2023). Once the retrieval has converged, the retrieved targets are used to simulate a final measurement spectrum which is referred to as the retrieved spectrum.

IMS also includes a number of parameters that can be optionally fixed in the retrieval.

The vertical profiles of CO₂, CH₄, and N₂O are by default represented by a monthly latitude-dependent climatology derived from the Monitoring Atmospheric Composition and Climate (MACC) greenhouse gas flux inversion reanalysis (Bergamaschi et al. 2013). The surface pressure is derived from ERA5 reanalysis data.

The iterations are based on the Levenberg-Marquardt method (Eq. 2.30), which was introduced in Section 2.2. In this method, the Levenberg-Marquardt parameter controls the magnitude of the state vector update and is initially set to 0.001 in this work. Within IMS, the retrieval is said to have converged when the cost function (J derived in Eq. 2.29) has reduced, is the lowest calculated so far, and the change in J is less than 1. If the cost increases but is below a variable threshold, the retrieval stops based on a low cost. If 10 iterations are reached and the criteria have not been met, the retrieval has not converged. These settings can be varied and have been optimised for use on IASI observations.

4.2.1 The State Vector

In this work, IMS is used to perform simultaneous retrievals of temperature (K), water vapour (ppmv), surface skin temperature (K), surface emissivity, cloud optical thickness (COT), cloud effective radius (CER; μm), and cloud top height (CTH; km). The details of the state vector components are tabulated in Table 4.1. The a-priori for temperature, water vapour, and surface skin temperature can be set to either a typical climatology or hourly ERA5 reanalysis data. In this work, the latter is used as a tighter constraint given the limited number of observations of far-infrared radiances. The corresponding a-priori covariance for temperature, water vapour and skin temperature is a 2-dimensional matrix derived from the differences between the zonal mean of ERA5 profiles for three days (17 April, 17 July, and 17 October 2013) and includes off-diagonals. These three days were selected by Richard Siddans (RAL Space) as they were originally used to determine a bias correction for the IASI Level 2 products by EUMETSAT.

The a-priori for surface emissivity is taken from the University of Wisconsin InfraRed land surface EMISsivity (UWIREMIS) atlas within RTTOVv12 (Seaman et al. 2008). For land, this uses the input latitude, longitude and snow fraction to determine the surface emissivity based

Table 4.1: A summary of the retrieval targets within the state vector in IMS. For the COT, the a-priori is stored in log form and the covariance represents the variation on the log of the COT.

Retrieval Target	A-priori	A-priori Covariance
Temperature (K) and water vapour (ppmv) profiles, and surface skin temperature (K)	Hourly ERA5	Hourly ERA5 from 3 days globally (17 Apr, Jul, Oct 2013)
Surface emissivity	UWIREMIS Atlas	UWIREMIS Atlas from 3 days globally (17 Apr, Jul, Oct 2013)
COT	$\ln(0.01)$	10^*
CER (μm)	30	10
CTH (km)	7	5

on an offline database. This offline database was compiled by fitting Moderate Resolution Imaging Spectroradiometer (MODIS) observed emissivities to measured emissivities of natural materials (Seeman et al. 2008). For sea, the emissivity is parameterised in terms of zenith angle, wind speed and skin temperature and calculated offline based on refractive indices from Hale & Querry (1973), and a wave slope model from Masuda (2006). The a-priori covariance for the surface emissivity is calculated by performing a direct calculation of the covariance between emissivity spectra from the UWIREMIS surface emissivity atlas for the same three days (17 April, 17 July, and 17 October 2013), and includes off-diagonals.

The a-priori and covariance for the cloud retrieval targets were selected to be representative of ice, water, or mixed phase clouds, and are 7 ± 5 km for CTH, and 30 ± 10 μm for CER. COT is stored in log form to prevent it from reducing below zero, and it is assumed there is minimal cloud at first, resulting in an a-priori of $\ln(0.01) \pm 10$. To date, these cloud parameters have been retrieved to reduce the impact of cloud contamination and bias on the retrieval of atmospheric gases (Trent et al. 2023, Dils et al. 2024). In this study, the goal is to retrieve the properties of the clouds themselves, and a more detailed description of this is provided in Chapter 6.

4.2.2 Radiative Transfer for TOVS (RTTOVv12)

The RTTOVv12 fast RT model is used as the forward model in the IMS scheme to simulate clear-sky and cloudy spectra using coefficients to increase operational speed (Saunders et al. 2018, 2017). It takes inputs of temperature, gaseous profiles, cloud fraction, CER, and cloud IWC at 101 fixed pressure levels from the surface to the top of the atmosphere.

For clear-sky radiances, the optical depths of each layer are parameterised by temperature and the gaseous profile into ‘clear sky’ coefficients. These coefficients are derived by performing a regression on a database of transmittances of atmospheric gases calculated from the top of the atmosphere to each fixed pressure level using LBLRTMv12.8 (Clough et al. 2005), for all the profiles in the European Centre for Medium-Range Weather Forecasts (ECMWF) diverse 83-profile dataset that was developed to capture a large variety of atmospheric states (Chevallier et al. 2006). The LBLRTM transmittances include the effects of 28 gas species, alongside continua (MT_CKDv3.2) due to water vapour (self- and foreign broadening), ozone, carbon dioxide, nitrogen, and oxygen (Saunders et al. 2017). They are adapted to match the instrument line shape of ~ 90 different satellite sensors, and so each set of regression coefficients is customised to a specific instrument (Saunders et al. 2018, 2017).

Cloudy radiances can be calculated using a number of different methods across different spectral regions. In this work, RTTOVv12 is set up to use the Chou approximation, where the optical depths associated with atmospheric emission and absorption (calculated using the clear-sky coefficients) are scaled to account for the cloud particle interactions (Chou et al. 1999). The Chou approximation uses three parameters: (i) the absorption coefficient, (ii) scattering coefficient, and (iii) a backscattering parameter which represents the backscattered radiation. These parameters have been calculated for the Baran et al. (2014) model, and for one of the Yang et al. (2013) and Baum et al. (2014) bulk optical property models. These cloud optical property models are parameterised into ‘cloudy’ coefficients in terms of IWC from $0.5\text{E-}5$ to 0.1 gm^{-3} and CER from $5\text{-}60\text{ }\mu\text{m}$. These models and the impact of the Chou approximation will be explored in more detail in Chapter 6.

4.2.3 Default Channel Selection

When performing the fit optimisation, $\mathbf{J}$ can be isolated to certain channels rather than being considered across the entire spectrum. Not all channels within a spectrum are sensitive to the retrieval targets, and so may not enhance the fitting process. Furthermore, a large instrument uncertainty reduces the information known about $\mathbf{y}$ (Rodgers 2000), and so excluding these channels could improve the final retrieval output. To select these channels, the DOFS or information content (described in Section 2.2.4) can be used to maximise the information available for the retrieval. Channel selections that have used either parameter have been shown to lead to comparable results (Rabier et al. 2002). Finally, fewer channels increase the speed of the retrieval framework, which is desirable for retrievals on hyperspectral images that contain a large number of spectra, or more complex cases.

As previously mentioned, IMS is currently used to perform retrievals on observations taken by IASI, and to do so, it uses a restricted set of channels. IASI covers the spectral range of $645\text{--}2760\text{ cm}^{-1}$ with a resolution of 0.5 cm^{-1} and sampling of 0.25 cm^{-1} . RTTOVv12 simulates IASI spectra across its full spectral range with a channel width of 0.25 cm^{-1} , totalling 8460 available channels to select from in the retrieval. The current IASI channel selection used

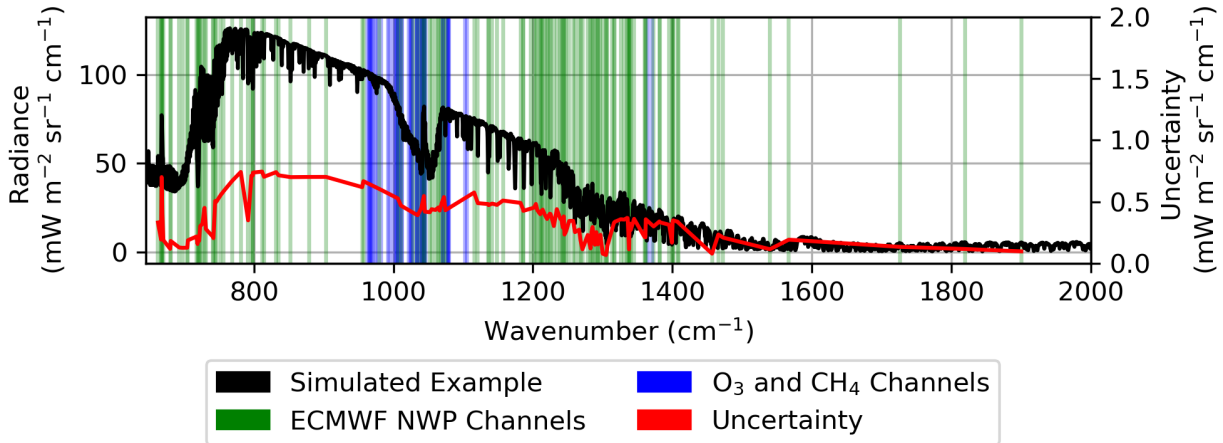


Figure 4.2: An example RTTOVv12 simulation of a spectrum measured by the IASI instrument overlaid with the 181 channels used within IMS to perform retrievals. The channels selected by ECMWF for use in Numerical Weather Prediction models are shown in green, and the additional channels selected for improving ozone and methane retrievals in IMS are shown in blue. The square-root-diagonal of $\mathbf{S}_y$ for IASI (Uncertainty) is also shown in red and is derived from residuals between IASI observations and RTTOVv12 simulations as described in Section 4.2.

within IMS is derived from the study in Collard (2007) that was used to select IASI channels that would be incorporated into Numerical Weather Prediction models. Channels were first filtered by noise and reliability, reducing the channels available for general retrievals to 300. From these, channels were then selected to maximise the DOFS across a diverse profile set. Final channels were then added or removed to enhance the retrieval outputs for these profiles on an ad-hoc basis, resulting in 138 channels for use in general retrievals from IASI observations to optimise retrievals of temperature, water vapour, and ozone (Collard 2007, Weston 2011). An additional 43 channels have also been included in IMS to further improve retrievals of O_3 and CH_4 by optimising the information content about each target. The final channels selected for retrievals from IASI observations in IMS are shown, in comparison to the corresponding square-root-diagonal of S_y , in Figure 4.2, totalling 181 channels within the mid-infrared.

4.3 Development Outline

The aim of this thesis is to extend the IMS retrieval scheme described in Section 4.2 to exploit far-infrared signals of water vapour and ice clouds. Figure 4.1 highlights in red the four components of IMS that are instrument-specific and must be altered to enable retrievals from far-infrared radiances: the coefficients, the spectrally dependent covariance matrices, and the channel selection.

For clarity, this chapter only focuses on clear-sky retrievals, and so the required adjustments to the cloudy coefficients are explored in Chapter 6. The clear-sky coefficients used by RTTOVv12, outlined in Section 4.2.2, have already been built for the FSI and were readily available on the RTTOV website. The first major adjustment is to build spectrally dependent covariance matrices that align with the FSI spectral grid and instrument specifications. The next adjustment is then to select the channels used for clear-sky retrievals from the FSI based on its target uncertainty. All of the modifications described in this chapter (integration of coefficients, development of covariance matrices, and channel optimisation) have been performed by the author of this thesis to enable IMS to perform retrievals on FSI observations.

To monitor the impact of changes made to IMS, it is first useful to assess how well IMS currently retrieves temperature and water vapour profiles from clear-sky spectra. The ability of IMS to retrieve temperature and water vapour from IASI observations is well documented (Trent et al. 2023). Therefore, to test these changes, I have constructed a set of clear-sky profiles that will be used to simulate RTTOVv12 spectra. With each adjustment to the retrieval framework, the retrieval bias and diagnostic parameters (Section 2.2.4) can be monitored to examine the impact of any changes made. The creation of these test cases is detailed in Section 4.4, with the retrievals from RTTOVv12 IASI simulated spectra using the original configuration contained within in Section 4.5. The extension is then detailed in Section 4.6 with the channel selection for the FSI configuration in Section 4.7.

4.4 Simulated Clear-Sky Test Cases

Given the limited number of observations of upwelling TOA radiances in the far-infrared region, preliminary testing has been performed using simulated test cases for the IASI and FSI configurations. The input or ‘true’ profiles are known for each test case, and as a result, the retrieved products can be compared against the truth without any introduction of uncertainty in the atmospheric state.

A number of existing artificial datasets have been crafted to encompass variable atmospheres, such as the ECMWF diverse 83-profile dataset, however, these have already been used in generating the RTTOVv12 coefficients and for the channel selection in Section 4.2.3 (Saunders et al. 2017). Reanalysis datasets, including but not limited to ERA5 and Copernicus Atmosphere Monitoring Service (CAMS) data, have also been used in the development of IMS and while the reliance of IMS on these datasets is very weak, they do not provide an independent basis for testing IMS (Siddans 2019). Therefore, a test set has been constructed from Modern-Era Retrospective analysis for Research and Applications v2 (MERRA-2) reanalysis data produced by NASA Global Modeling and Assimilation Office (Gelaro et al. 2017). MERRA-2 has been shown to perform comparably to ERA5 when assessed against observations and is an independent base for this test set (Jiang et al. 2015, Arshad et al. 2021,

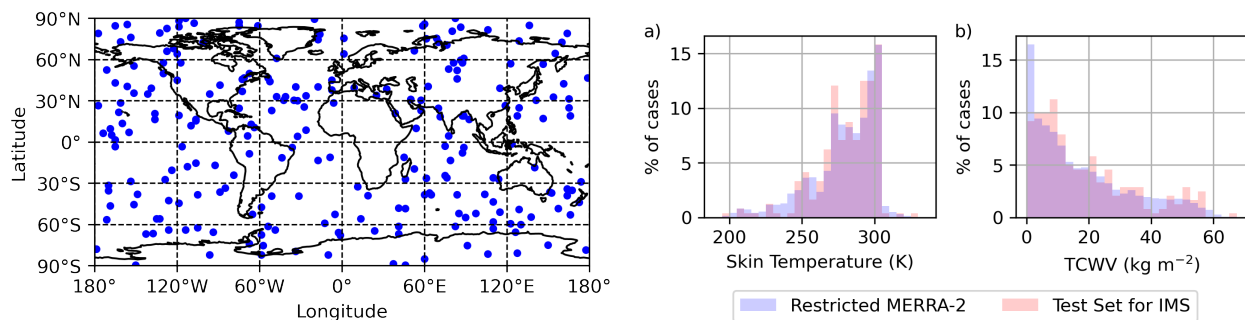


Figure 4.3: (a) The spatial distribution of the 240 cases selected for testing IMS. Profiles were derived from MERRA-2. Tropical Cases: 70. Mid-Latitude Cases: 95. Arctic Cases: 75. The percentage of total cases with each (b) skin temperature, and (c) total column water vapour (TCWV) in the MERRA-2 reanalysis dataset for one day in March, June, September, and December in 2019 and used in the test dataset, shown in blue and red, respectively. Overlapping regions are shown in purple where the same proportion of cases is present in the test set and MERRA-2 reanalysis dataset.

Huang et al. 2021, Johnston et al. 2021).

Instantaneous 3-hourly MERRA-2 reanalysis data were used for temperature, water vapour, and ozone vertical profiles (Global Modeling and Assimilation Office & Pawson 2015b). Ozone is not a retrieval target in this work and is fixed to the ‘true’ profile in each retrieval, but it has been chosen to vary across the test cases given its strong absorption in the mid-infrared. Instantaneous hourly MERRA-2 reanalysis data were used for skin temperatures and surface pressures (Global Modeling and Assimilation Office & Pawson 2015a).

The MERRA-2 data were first restricted to the 1st of March, June, September, and December 2019 to cover a broad range of seasons and conditions. From these data, 240 test cases were selected to capture variations in temperature and water vapour near the surface and in the upper troposphere. The spatial distribution of test cases can be seen in Figure 4.3a, and the distribution of skin temperature and total column water vapour in Figure 4.3b and c. The selected test cases, shown in red, capture the upper and lower limits of the restricted MERRA-2 data, shown in blue, for skin temperature and total column water vapour (TCWV), however the distribution of test cases have a slightly smaller proportion of cases with a TCWV below 2.5 kgm⁻². Overall, the finalised test cases demonstrate a comparable spread relative to the initially selected MERRA-2 days.

The US 1976 Standard profiles for CO₂, CH₄, and N₂O were scaled to match NOAA flask

measurements averaged from 2019 (Saunders et al. 2017, Dlugokencky et al. 2021), and have been kept constant across all the test cases. Realistic surface emissivities in the mid-infrared were selected based on their MODIS surface type from the UWIREMIS atlas (Platnick et al. 2003, Seeman et al. 2008).

4.5 Evaluating the Current Ability of IMS

Retrievals of temperature, water vapour, surface skin temperature, and surface emissivity have been performed using the IASI covariances and channels outlined in Section 4.2.3 from RTTOVv12 simulated IASI spectra based on the profiles described in Section 4.4. This is referred to as the IASI configuration. Figure 4.4 summarises the difference between the ‘true’ and retrieved state in terms of the mean retrieval bias for all 240 cases and has been divided into latitudinal regions.

Overall, the retrieved temperature and water vapour profiles generally have a smaller bias to the ‘true’ profiles in comparison to the a-priori estimates, regardless of their initial direction. The performance of the IASI configuration shows comparable retrieval biases to those observed in Trent et al. (2023) from IASI observations, with a mean bias within 12% and 0.3 K for water vapour and temperature retrievals, respectively, in comparison to the 10% and 1 K observed in Trent et al. (2023) against radiosonde measurements. These are driven by inherent biases between the a-priori, taken from ERA5, and the MERRA-2 profiles used to simulate the test cases (Johnston et al. 2021).

The retrieved water vapour profiles show a wet bias to the ‘true’ profiles, with larger biases observed in the mid- to upper troposphere as observed in Trent et al. (2023) (Figure 4.4c and d). While the temperature retrievals perform better with a mean temperature bias less than 0.5 K throughout the entire profile, Figure 4.4g and h show increases of up to 0.4 K in the temperature bias in comparison to the a-priori. High-latitude cases were noted to have a poorer performance in Trent et al. (2023) and the ESD associated with both the water vapour retrievals in the polar regions (Figure 4.4b and f) is up to 21 % larger than those seen in the mid-latitudes and tropical regions. However, the ESD associated with the temperature

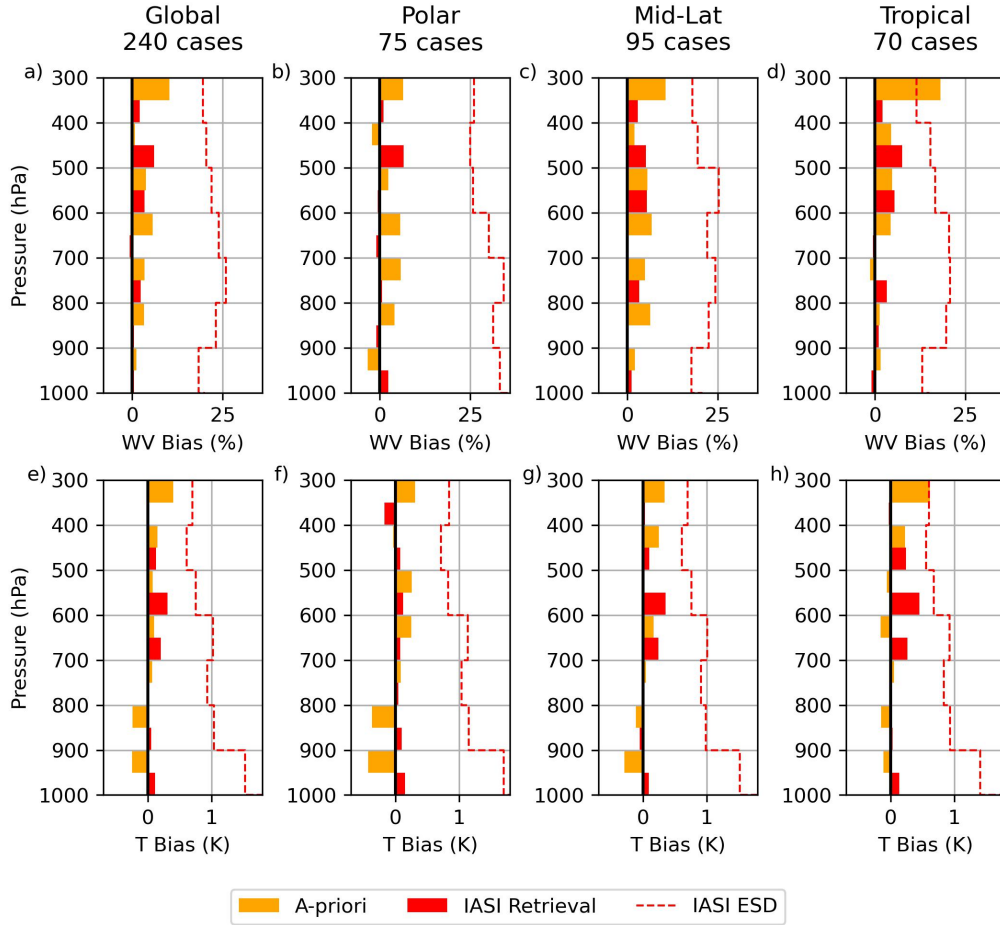


Figure 4.4: The median (a-d) water vapour (WV) and (e-h) temperature (T) bias of the retrieved profile to the ‘true’ state across the testing cases for the optimised IASI configuration in red, in comparison to the a-priori bias to the ‘true’ state in orange. Biases are evaluated between 300 and 1000 hPa in 100 hPa bins as in Trent et al. (2023) and shown for (Global) all 240 cases as well as in separate latitudinal regions of (Polar) 60 to 90 degrees N/S, (Mid-Latitudes) 30 to 60 degrees N/S, and (Tropical) 30 N to 30 S. The water vapour percentage bias is calculated from absolute values in ppmv. Dashed lines show the median estimated standard deviation (ESD) for a single retrieval for the test cases in each bin. For water vapour this is plotted with an offset of -10 % for clarity.

retrieval in the polar region is only 0.2 K larger than what is observed in the mid-latitude and tropical regions.

Table 4.2 summarises parameters that will be used to compare the IASI retrieval to the configurations constructed in the following sections. The temperature DOFS are almost twice as large as the water vapour DOFS for water vapour (Table 4.2) due to the sensitivity of the CO₂ wings to the temperature profile. The skin temperature and surface emissivity are summarised in these parameters, and both show that the skin temperature is, on average, captured to within 0.1 K and the surface emissivity within 0.002.

Table 4.2: A summary of the median and median absolute deviation in diagnostic parameters for the retrievals from the 240 test cases using the IASI configuration. The absolute residual skin temperature and surface emissivity residual were calculated by finding the median of the absolute difference between the retrieved and 'true' values. The remainder of the diagnostic parameters are described in Section 2.2 with definitions in Table A2.

Diagnostic Parameter	IASI Retrieval
Water Vapour DOFS	5.0 ± 0.8
Temperature DOFS	9.9 ± 0.4
J_y Per Channel	0.06 ± 0.04
J_x	7 ± 2
Absolute Residual Skin Temperature (K)	0.08 ± 0.05
Absolute Surface Emissivity Residual	0.0012 ± 0.0001

To conclude, IMS has now been evaluated on 240 IASI simulated spectra covering a diverse range of temperature and water vapour profiles, respectively. Results are consistent with prior work comparing IMS retrievals from IASI observed spectra to radiosonde measurements (Trent et al. 2023), with final maximum ensemble mean biases of 0.3 K and 12 % for temperature and water vapour profiles. The next step in this work is to extend IMS into the far-infrared. The same test cases will be used to simulate FSI spectra, allowing a direct comparison between retrievals from simulated IASI and FSI spectra, as well as an assessment of the impact of including the far-infrared channels for the expected FSI configuration.

4.6 Technical Extension for the FORUM Sounding Instrument

This section describes the inclusion of the FSI clear-sky regression coefficients and FSI-compatible covariance matrices in IMS, and the testing IMS has undergone using the test set described in Section 4.4. The RTTOVv12 FSI clear-sky coefficients were provided by the RTTOV team. I have performed all of the other work within this section.

4.6.1 Using the FSI Clear-Sky Regression Coefficients

The clear-sky coefficients used to simulate spectra in RTTOVv12 have already been built for the FSI and are available on the RTTOV website. These can be easily integrated into the retrieval framework. The first thing to consider when introducing these into the retrieval framework is the difference in spectral characteristics to IASI. The RTTOVv12 FSI radiances cover the full FSI spectral range at a sampling of 0.3 cm^{-1} and have the strong Norton-Beer apodisation applied. This results in a total of 5000 channels and a spectral resolution of 0.6 cm^{-1} . In comparison, RTTOVv12 simulates IASI radiances between $645 - 2760 \text{ cm}^{-1}$ with a sampling of 0.25 cm^{-1} and resolution of 0.5 cm^{-1} totalling 8460 channels. As outlined in Section 4.2.3, only 181 of these channels are currently used in IMS for retrievals from IASI observations. Three of these channels are outside the FSI spectral range: 1726.0 cm^{-1} , 1819.5 cm^{-1} , and 1900.0 cm^{-1} . All of these overlap with the water vapour vibration-rotation band centred at 1595 cm^{-1} . However, they have a limited sensitivity to water vapour and were selected to improve O_3 and CO_2 retrievals.

As a first step, the retrievals using the IASI configuration described in Section 4.5 have been repeated without the channels that exceed the FSI spectral range, resulting in 178 channels used within the retrieval. This will be referred to as the IASI 178 retrieval, and the results are shown in Figure 4.5. These show that the removal of the three channels has a minimal impact on the retrieval bias with no change to the parameters in Table 4.2.

The next step is to introduce the FSI clear-sky coefficients into IMS and perform retrievals on the testing set of the 240 cases that have been simulated using the FSI coefficients. As described, the channel width and position differ between the FSI and IASI, so as a first iteration, the closest FSI channel to each of the 178 IASI channels has been used in the retrieval (with the retrieval termed FSI 178). The average offset between an FSI channel and its IASI counterpart was 0.05 cm^{-1} . The values of the spectrally dependent covariances were taken for each of the original IASI channels, and no interpolation was performed to correct for the difference in channel position.

Figure 4.5 shows comparable retrieval biases between the IASI 178 and FSI 178 configurations within their respective ESD values. Below 500 hPa, the biases associated with the

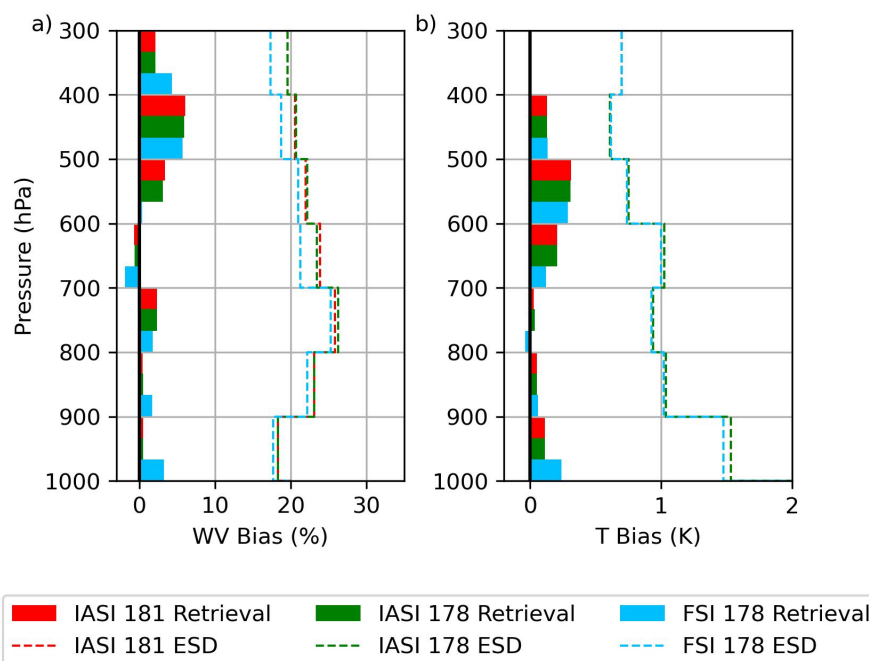


Figure 4.5: The median (a) water vapour (WV) and (b) temperature (T) bias of the retrieved profile to the ‘true’ state across the testing cases for three different retrieval configurations. The retrievals using the IASI configuration and 181 channels is shown in red and is the same as in Figure 4.4. The retrievals using the IASI configuration with 178 channels is shown in green. The retrievals using the FSI configuration with the FSI channels closest to the 178 IASI channels is shown in light blue. The water vapour percentage bias is calculated from absolute values in ppmv. Dashed lines show the median estimated standard deviation (ESD) for a single retrieval for the test cases in each bin. For water vapour this is plotted with an offset of -10% for clarity.

FSI 178 configuration increase towards the surface for both temperature (0.2 K) and water vapour (4 %). There is a slight increase in the absolute residual skin temperature of 0.01 K accompanied by a slight increase in the absolute skin emissivity residual of 0.001. It is important to note that this channel selection was optimised for the IASI instrument line shape. Hence, the position and sensitivity of absorption features will differ from those observed by the FSI. Furthermore, given the wider channels associated with the FSI 178 configuration, a worse retrieval is expected as there is a loss of spectral information associated with each channel. Both of these issues will be explored in the remainder of this chapter.

4.6.2 Building the New Covariance Matrices

The next stage of this development is to extend the spectrally dependent covariance matrices contained within IMS. The a-priori covariance for the surface emissivity and measurement

covariance are both spectrally dependent and so must be tailored to the specifications of the FSI defined in RTTOVv12. They are currently configured for the mid-infrared and microwave spectral regions with spectral sampling and ranges for instruments on board the MetOp satellites.

Measurement Error Covariance

As discussed in Chapter 2.2, the measurement covariance is used to represent the uncertainty in the observed spectra and forward model. For IASI, the residuals between observations and RTTOVv12 simulations are used to constrain the retrieval. For the FSI, the measurement covariance has been built using the apodised target NESR and target ARA, which are shown in Figure 4.6. The target ARA is 0.1 K between 300 and 1100 cm^{-1} , 0.2 K between 200 and 300 cm^{-1} and 1100 and 1300 cm^{-1} , and 1K elsewhere. As the ARA is defined in brightness temperature, it is converted into radiance units for each individual spectrum, and the average is shown in Figure 4.6a for each profile in the ECMWF diverse 83-profile dataset (Chevallier et al. 2006).

The target NESR and ARA for the FSI have been combined to produce a correlated measurement covariance matrix. It has been separated into the NESR and ARA components in Figure 4.6b and c with the full matrix in Figure 4.6d. The apodised NESR includes four sets of non-zero off-diagonals due to the thin instrument line shape, from the Norton-Beer strong apodisation, and is the dominant contributor to the measurement covariance below 1300 cm^{-1} and above 1450 cm^{-1} . It is assumed that the ARA covariance is fully correlated as it represents a systematic bias in the radiances, and so it has contributions off the leading diagonal of up to $0.5 [\text{mWm}^{-2}\text{sr}^{-1}/\text{cm}^{-1}]^2$ below 100 cm^{-1} and above 1300 cm^{-1} .

As can be deduced from Figure 4.6a, the FSI measurement covariance is between 0.08 and $0.37 [\text{mWm}^{-2}\text{sr}^{-1}/\text{cm}^{-1}]^2$ smaller than its IASI counterpart between 750 and 1200 cm^{-1} . At wavenumbers above 1300 cm^{-1} , the FSI measurement covariance increases, reaching a difference of $0.9 [\text{mWm}^{-2}\text{sr}^{-1}/\text{cm}^{-1}]^2$ at 1600 cm^{-1} .

Figure 4.7 shows the retrieval biases associated with retrievals using the FSI 178 configuration (previously described) that uses the IASI measurement covariance, and the FSI

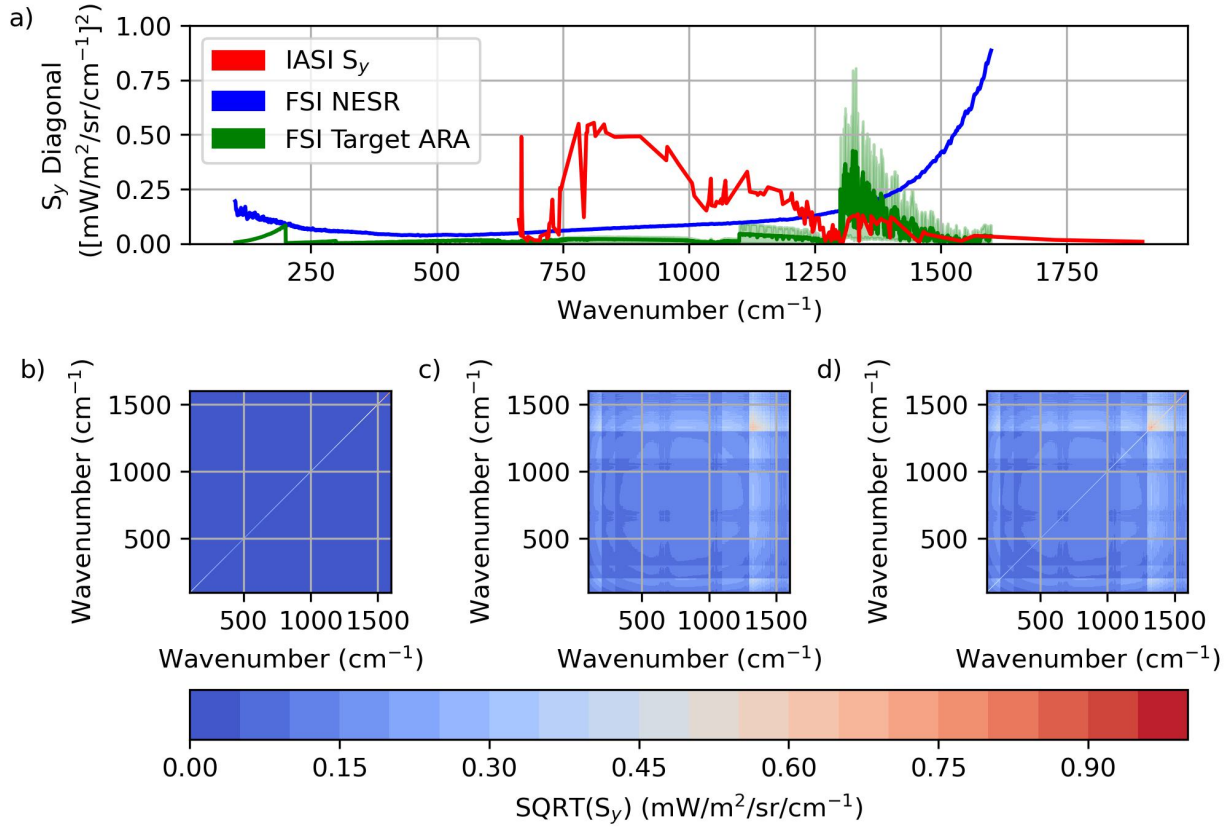


Figure 4.6: (a) The main diagonal of the IASI measurement covariance in red and the main diagonal of the FSI measurement covariance separated into components from the noise-equivalent-spectral-radiance (NESR) in blue and the target absolute radiometric accuracy (ARA) in green. As the ARA is provided in brightness temperature, it has been converted into radiance units for each of the ECMWF 83 profiles. The median is shown here, with the shading representing the minimum and maximum values. The square-root of the measurement covariances for the (b) NESR, (c) ARA, and (d) final combined matrix are also shown.

178 S_y configuration, which now uses the FSI measurement covariance in each of the 178 channels. As all of the channels used were selected for IASI and are greater than 645 cm^{-1} , the additional information associated with the far-infrared is not included. Information regarding the water vapour profile is coming from the water vapour vibration-rotation band centred at 1595 cm^{-1} , where the FSI uncertainty is larger than its IASI counterpart. A larger measurement covariance suggests less information is available about the ‘true’ state and reduces the quality of the retrieval products. This is most clearly reflected through the increase in the ESD of up to 12 % and 0.3 K for water vapour and temperature, respectively. However, the median biases themselves show a minimal increase of 2 % and 0.3 K for water vapour and temperature, respectively, with the use of the FSI measurement covariance. Furthermore,

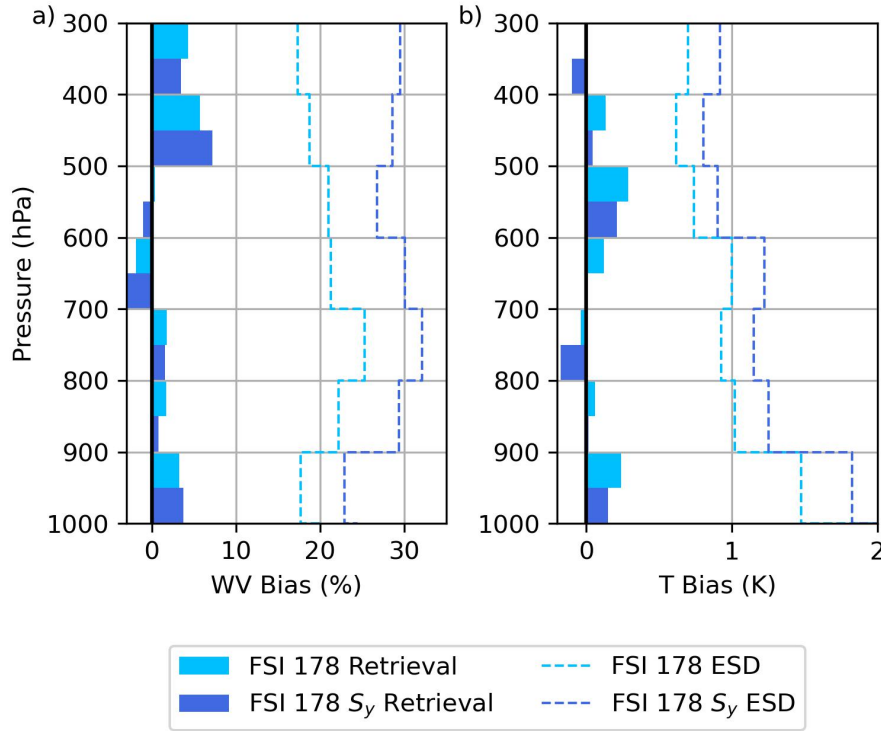


Figure 4.7: The median (a) water vapour (WV) and (b) temperature (T) bias of the retrieved profile relative to the ‘true’ state across the testing cases for three different retrieval configurations. The retrievals using the FSI configuration with the FSI channels closest to the 178 IASI channels is shown in light blue (FSI 178). The retrievals using the FSI configuration with the FSI channels and measurement covariance closest to the 178 IASI channels is shown in blue (FSI 178 S_y). The water vapour percentage bias is calculated from absolute values in ppmv. Dashed lines show the median estimated standard deviation (ESD) for a single retrieval for the test cases in each bin. For water vapour this is plotted with an offset of - 10% for clarity.

following Eq. 2.29, this larger measurement covariance results in a smaller measurement cost for the FSI 178 S_y configuration of 0.023 ± 0.008 . The state cost has also significantly reduced by ~ 2 , suggesting that the retrieved state has remained close to the a-priori as the retrieval can converge more easily. No significant impact is observed on the retrieved surface properties.

A-priori Covariance for Surface Emissivity

The next covariance that must be adjusted is the a-priori covariance for surface emissivity. The surface emissivity is retrieved as it has been shown to improve the agreement between retrievals of lower tropospheric water vapour and ECMWF analyses, as well as reducing cloud contamination (Trent et al. 2023). The a-priori covariance for surface emissivity is only

defined for the spectral ranges of the instruments available within IMS, and so currently does not extend into the far-infrared.

As described in Section 4.2, the a-priori covariance for surface emissivity was calculated separately for sea and land surface types before being combined into one covariance matrix. The covariance of the sea surface emissivity was derived from a database of refractive indices dependent on zenith angle, wind speed, and skin temperature, and a wave slope model (Hale & Querry 1973, Masuda et al. 1988). These models extend into the far-infrared, and so have been used in this work to simulate ~ 20000 surface emissivities, that cover the FSI spectral range, by varying the zenith angle ($0-85^\circ$), wind speed ($0-100 \text{ ms}^{-1}$), and skin temperature ($211-318 \text{ K}$) to cover the extent of possible inputs into RTTOVv12. The covariance of this dataset was used to find the a-priori covariance for sea surface emissivity.

For land cases, the surface emissivity produced by the UWIREMIS atlas is fixed in the far-infrared, and so the covariance was instead found using the Huang et al. (2016) emissivity database (Figure 4.8) that is theoretical in the far-infrared. Currently, there is a large uncertainty associated with the surface emissivity of different land types in the far-infrared (Ben-Yami et al. 2022), and so to provide a loose constraint on the surface emissivity retrieval, the covariance has been considered across the different surface types without a weighting for global coverage. This database has also been used to define the far-infrared surface emissivity of the test cases over land described in Section 4.4.

The existing a-priori covariance for surface emissivity reaches 645 cm^{-1} , and so this was first interpolated onto the FSI channels. The land and sea covariances calculated in this section were then added in quadrature and offset by 0.0005 to enable a smooth transition between the new (less than 645 cm^{-1}) and existing (greater than 645 cm^{-1}) a-priori covariance matrices. The updated a-priori covariance for surface emissivity is shown in Figure 4.9 and includes off-diagonals representing correlations between different channels. Greater variations are observed in the covariance matrix between $400-600 \text{ cm}^{-1}$ and in the atmospheric window that can be attributed to the lower values of desert surface emissivity in these spectral regions (Huang et al. 2016). The impact of this covariance matrix on this retrieval will be considered in the following section, where channels in the far-infrared are introduced.

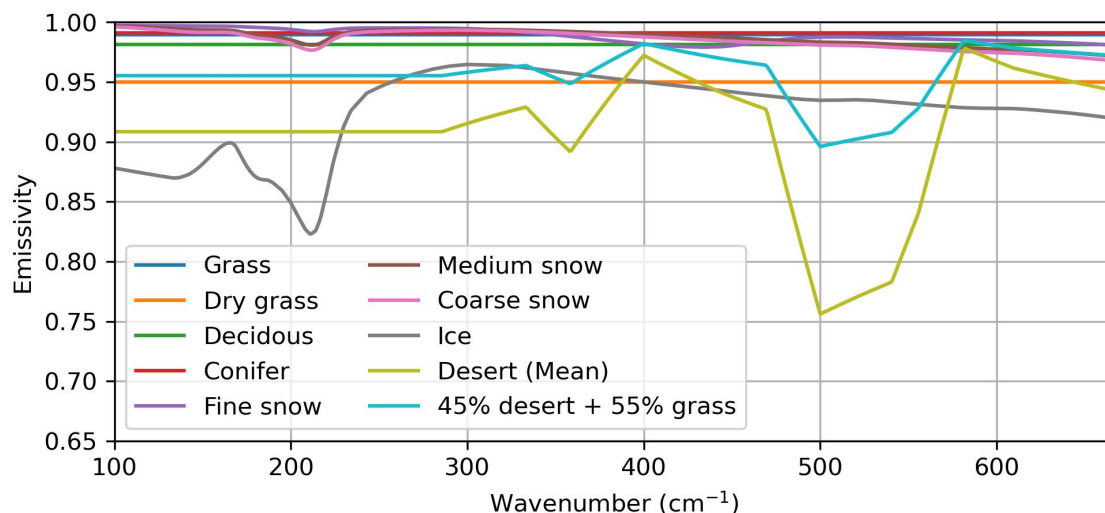


Figure 4.8: The different far-infrared surface emissivities contained within the Huang et al. (2016) dataset prescribed for different surface types. The Desert (Mean) surface emissivity corresponds to the average of the different desert surface types contained within the model that have different silt sizes. The surface emissivity for grass is partially obscured by the surface emissivity for conifers. Uncertainties for these emissivities are not provided with this database and the accuracy of each surface emissivity in the far-infrared has not been thoroughly validated.

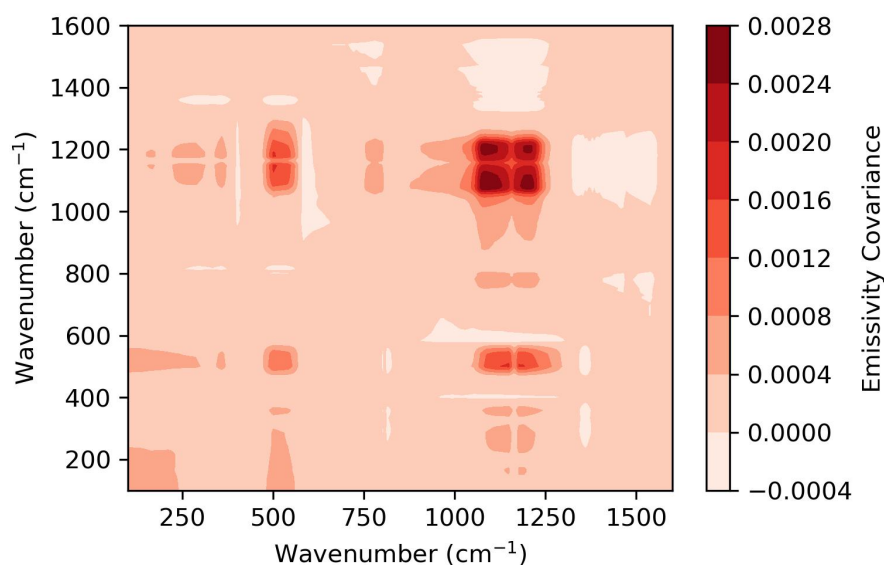


Figure 4.9: The a-priori covariance for the surface emissivity for the FSI configuration. Above 645 cm^{-1} , the original IMS covariance has been interpolated onto the FSI spectral grid. Below 645 cm^{-1} , the covariance has been derived from a combination of the (sea) UWIREMIS atlas and the (land) Huang et al. (2016) emissivity database.

4.7 Optimising for Clear-Sky Retrievals from FSI Radiances

This section describes the selection of channels used to perform retrievals for the FSI configuration. A similar analysis to the IASI channel selection described in Section 4.2.3 is performed here to optimise the temperature and water vapour retrievals by using the information content to rank and filter channels. The parameter for information content (Eq. 2.49) is dependent on the sensitivity of the retrieval to the measurement and a-priori covariance, and so it can be used to filter out channels with a greater uncertainty and weaker sensitivity to the retrieval products. In this case, the a-priori covariance is a weak constraint, and so the measurement covariance plays a more significant role in the channel selection.

First, the 5000 channels available with the FSI configuration were ranked based on their contribution to the total available information content for each retrieval from the ECMWF diverse 83-profile set (Chevallier et al. 2006). Iterative tests were then performed to assess how many channels were required before the improvement in information content plateaued. These tests were performed by including between 100 and 1500 channels, beginning from the

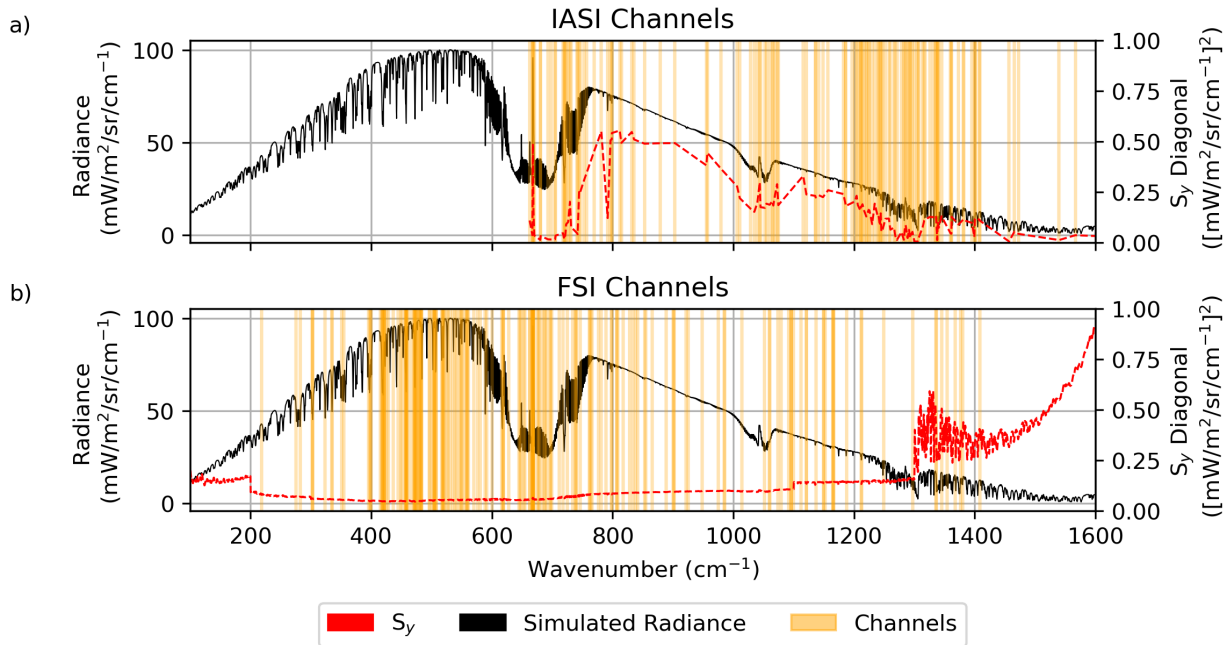


Figure 4.10: An example upwelling FSI spectrum simulated using RTTOVv12 overplotted with channels selected for (a) IASI and (b) FSI (FORUM) retrievals. The main diagonal of the measurement covariance $\mathbf{S}_y$ is shown in red in both cases. For IASI, $\mathbf{S}_y$ is derived from the differences between simulations and observations. For FSI, $\mathbf{S}_y$ is based on a combination of the target NESR and ARA and is the same as the green line in Figure 4.6a.

channels with the highest information content. It was found that the improvements in the total information content, as well as DOFS for temperature and water vapour, plateaued for the FSI configuration at 200 channels. No notable improvements in the DOFS or information content were found above 200 channels. Therefore, these 200 channels have been selected for use in IMS for the FSI configuration and are shown in Figure 4.10b. These capture $\sim 66\%$ of the total available information content, with the remaining channels each contributing at most 0.1%. An example of the vertical sensitivity of seven of the selected channels is shown in Figure 4.11 for one of the mid-latitude profiles in the ECMWF diverse 83-profile set.

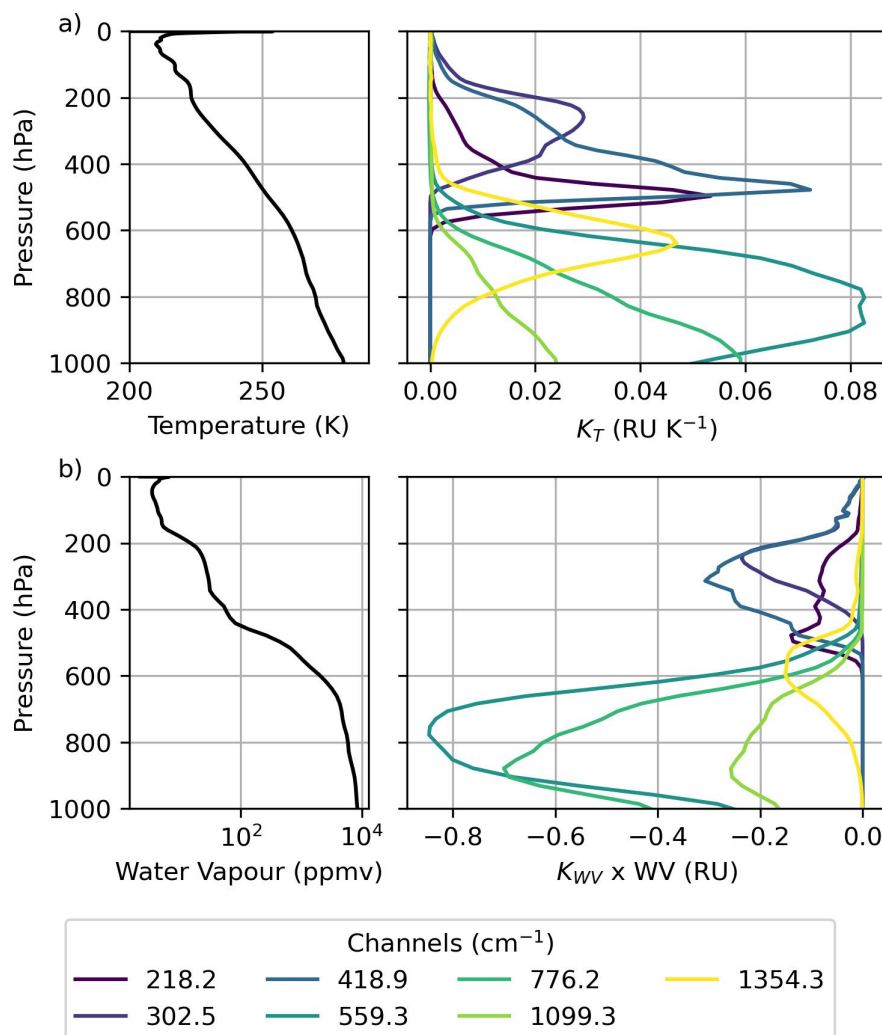


Figure 4.11: Example Jacobians of (a) temperature (K_T) and (b) water vapour (K_{WV}) for 7 of the 200 channels used in the retrieval. The Jacobians were calculated based on a mid-latitude profile (profile 42) in the ECMWF diverse-83 profile set using RTTOVv12. The temperature and water vapour (WV) profiles are shown in black. The Jacobians are given in terms of radiance units (RU), which are $\text{mW m}^{-2} \text{sr}^{-1} / \text{cm}^{-1}$. For clarity, the Jacobian for water vapour has been scaled by the water vapour profile.

Unlike the IASI channels (shown for comparison in Figure 4.10a), no channels have been selected at wavenumbers greater than 1400 cm^{-1} . Similarly, no channels below 200 cm^{-1} have been selected. A markedly reduced percentage of channels are located between $1000\text{-}1400 \text{ cm}^{-1}$ in comparison to the IASI selection as the noise characteristics associated with the FSI have effectively shifted the locations of peak information to the range $400\text{-}700 \text{ cm}^{-1}$ for water vapour, and information relating to ozone (and other trace gases) has not been prioritised.

Retrievals on RTTOVv12 simulations of all the 240 clear-sky test cases outlined in Section 4.4 have been performed for both the IASI and optimised FSI configurations. Table 4.3 summarises the evaluation metrics for retrievals performed using both the IASI and FSI configurations. Broadly speaking, the performance of the two configurations is similar, with each metric agreeing within the associated uncertainty seen across the test cases. The FSI configuration does appear to have a slightly increased state cost, indicative of a greater divergence from the a-priori state, which iterative tests suggest is linked to the addition of information from the far-infrared channels and the channel optimisation in line with findings in Merrelli & Turner (2012). It is worth recalling that the FSI measurement covariance consists solely of the estimated FSI NESR and target calibration uncertainty, while the IASI measurement covariance implicitly also includes forward model error.

The average DOFS for water vapour and temperature were also greater for the FSI configuration in this comparison, with observed improvements primarily in the upper troposphere. This is shown in the median cumulative degrees of freedom for signal (CDOFS) in Figure 4.12 which is indicative of the vertical resolution of the retrieved profile. The FSI configuration (blue) has a faster rate of increase with pressure in the CDOFS between 200

Table 4.3: A summary of the median and median absolute deviation in diagnostic parameters for retrievals from the 240 test cases using the IASI and FSI configurations.

Diagnostic Parameter	IASI	FSI
Water Vapour DOFS	5.0 ± 0.8	5.6 ± 0.8
Temperature DOFS	9.9 ± 0.4	10.4 ± 0.3
J_y Per Channel	0.06 ± 0.04	0.04 ± 0.01
J_x	7 ± 2	9 ± 3

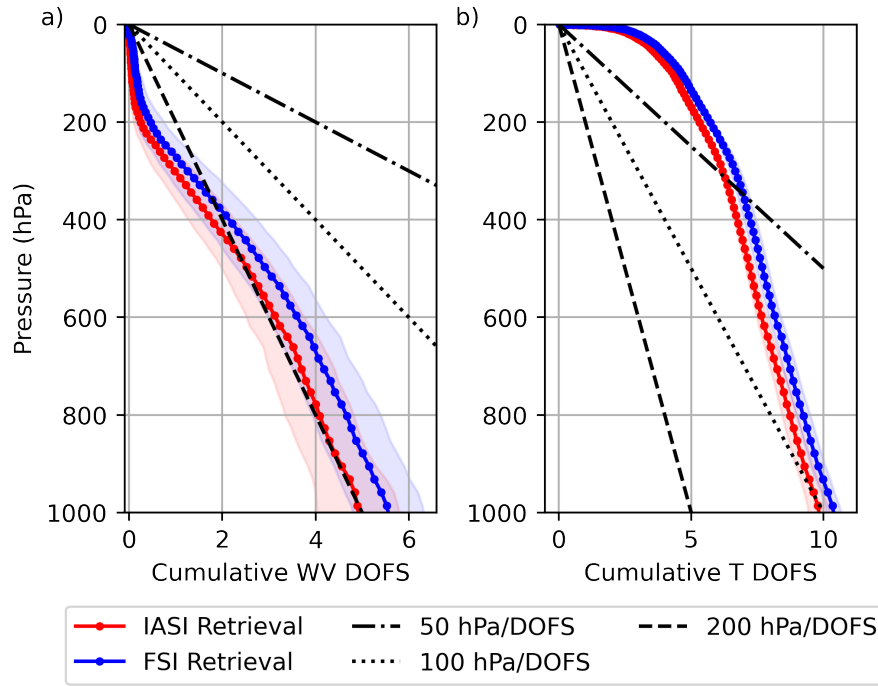


Figure 4.12: The median cumulative degrees of freedom for signal (CDOFS) for (a) water vapour (WV) and (b) temperature (T) averaged across all 240 cases for the IASI and FSI optimised configurations in red and blue, respectively. Shaded regions represent the median absolute deviation. This shows how the distribution of the vertical information can be related to the vertical resolution of the retrieved profile.

and 400 hPa for both temperature and water vapour. This suggests a slightly higher vertical resolution for the FSI configuration in the upper troposphere in comparison to the IASI configuration. Below 600 hPa, both configurations have a CDOFS that aligns with a vertical resolution of approximately 200 hPa/DOFS for water vapour and temperature.

The median retrieval biases for both configurations are presented in Figure 4.13. The FSI and IASI retrieval configurations perform comparably throughout the vertical, with biases within 2 % and 0.2 K of each other for water vapour and temperature, respectively. Generally, the FSI configuration has a lower median water vapour bias of up to 2%. Between 600 and 700 hPa, the FSI retrieval does not perform as well, with its water vapour bias reaching up to -2.5%, however this remains within the median ESD of the retrieval. Noticeably, the ESD associated with the FSI configuration for water vapour is up to 7% smaller throughout the vertical profile than its IASI counterpart, suggesting a higher confidence in the retrieved state. However, this is not observed in the temperature retrievals, with the ESD comparable between configurations.

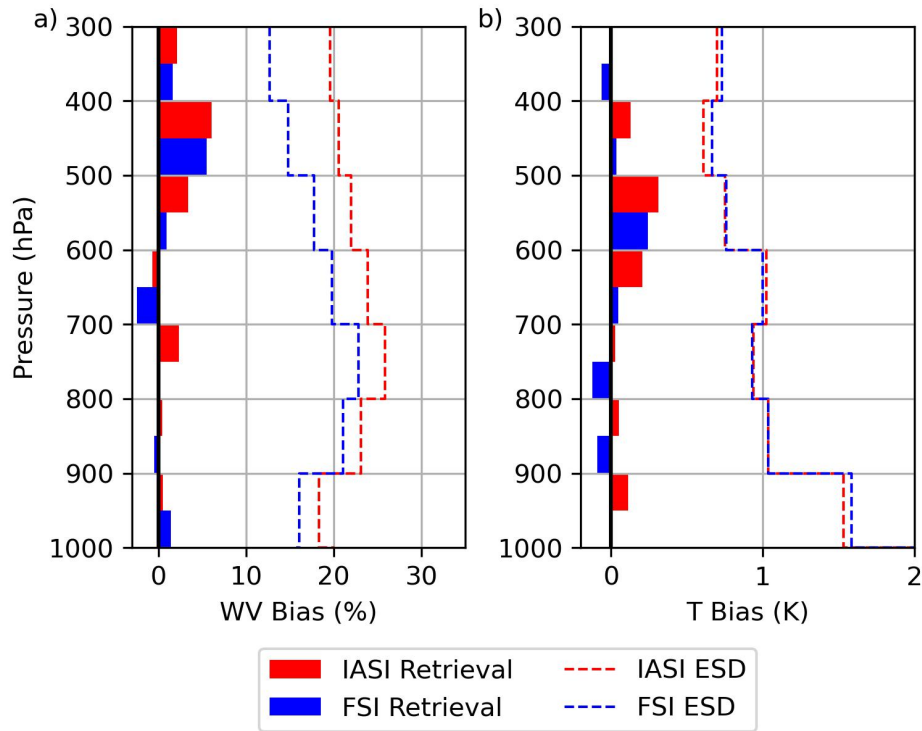


Figure 4.13: The median (a) water vapour (WV) and (b) temperature (T) bias of the retrieved profile to the ‘true’ state across the testing cases for the optimised IASI and FSI (FORUM) configurations in red and blue, respectively. Biases are evaluated between 300 and 1000 hPa in 100 hPa bins as in Trent et al. (2023). The water vapour percentage bias is calculated from absolute values in ppmv. Dashed lines show the median estimated standard deviation (ESD) for a single retrieval for the test cases in each bin. For water vapour, this is plotted with an offset of - 10% for clarity.

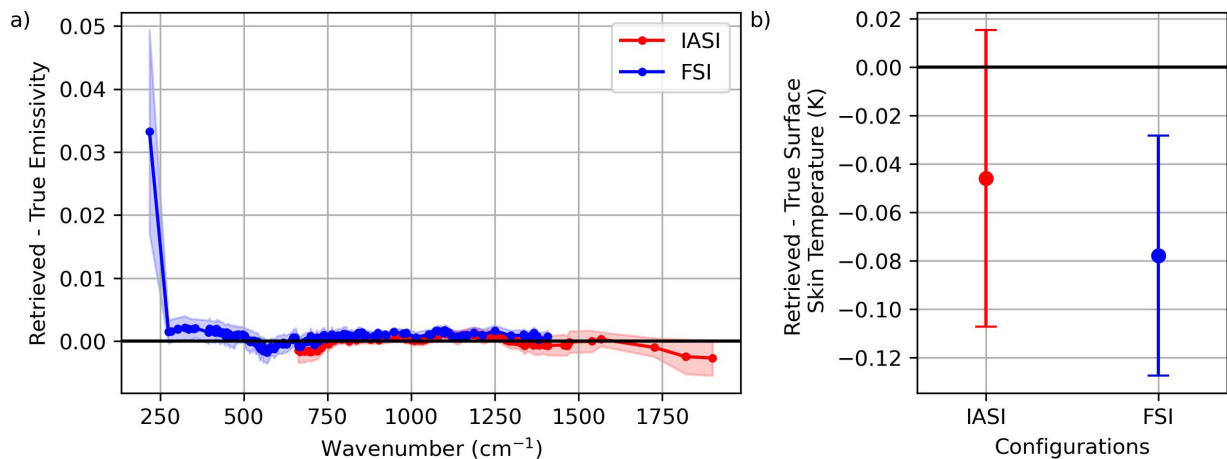


Figure 4.14: The median residual between the retrieved and ‘true’ (a) surface emissivity and (b) surface skin temperature for the IASI (red) and FSI (blue) configurations across the 240 cases. The shaded regions and error bars represent the median absolute deviation around this residual.

Figure 4.14 shows the median surface emissivity and skin temperature retrieved from the 240 test simulations using the IASI and FSI configurations. Figure 4.14a shows that in both cases, the retrieved emissivity is within 0.01 of the ‘true’ value above 250 cm^{-1} . Larger median residuals are observed below 250 cm^{-1} due to a lack of sensitivity in this region to the surface emissivity. Figure 4.14b shows that both configurations have a similar median retrieval bias for the skin temperature of approximately -0.08 K .

4.8 Conclusions

This chapter presents the extension of the Infrared Microwave Sounding retrieval scheme into the far-infrared in preparation for the upcoming FORUM mission. Retrievals of temperature and water vapour have been evaluated on simulated FORUM Sounding Instrument clear-sky radiances in preparation for Chapter 5. Comparisons are made to the ‘true’ profiles used to generate the simulations.

For the new FSI configuration, the measurement covariance is based on the expected apodised NESR and target ARA for the FORUM Sounding Instrument. An information content analysis was performed to optimise the channels used in the retrieval, resulting in the selection of 200 channels. The highest channel density was seen in the $400\text{--}600\text{ cm}^{-1}$ region due to the sensitivity of radiances to water vapour and temperature coupled with the relatively low target instrument uncertainty in this region.

The performance of IMS was first assessed using 240 RTTOVv12 simulations of top-of-atmosphere radiances based on MERRA-2 reanalysis profiles. Retrievals using the FSI configuration had a tighter measurement covariance than the IASI configuration below $\sim 1200\text{ cm}^{-1}$. Both configurations demonstrated comparable retrieval biases. However, slight improvements in the FSI median retrieval bias were observed in the mid- to upper troposphere for water vapour, with a reduced median uncertainty of at most 7 %, and an increase of ~ 1 DOFS for both water vapour and temperature due to additional information in the upper troposphere. The goal of the comparison is not to assess which instrument is better in terms of their retrieval performance, but rather a sanity check of the new, extended IMS

code. In particular, it is worth noting that in this comparison the measurement covariance used for IASI is not the quoted instrument NESR but instead implicitly includes the effects of instrumental and forward model error. For the FSI, the instrumental error has been approximated using the target instrument specifications, and so it is likely that the comparison overstates the improvement that FORUM will bring relative to IASI observations.

In summary, IMS can now perform retrievals on FSI radiances. The next stage of this work is to apply this retrieval scheme to observations of upwelling far- and mid-infrared radiances taken at aircraft-level.

Chapter 5

Application to Clear-Sky Observations from the Upper Troposphere

Key Points

- This chapter describes the application of the extended Infrared Microwave Sounding retrieval scheme to airborne observations of upwelling far- and mid-infrared radiances in clear-sky conditions.
- The channel selection and measurement covariance have been reconfigured for the FORUM-aircraft observations described in Chapter 3. Initial testing has been performed on radiances simulated using the profiles outlined in Chapter 4 at the observing altitude of the aircraft. Results showed a comparable retrieval bias to its TOA counterpart from Chapter 4.
- Retrievals have then been performed on eight observed spectra, and good agreement was found to in-situ measurements of temperature and water vapour; however, some limitations in the retrieved temperature profile were seen.

5.1 Introduction

The next stage of this thesis is to assess the ability of the IMS retrieval framework on aircraft-level observations of upwelling radiances in the far- and mid-infrared. This will highlight limitations in the forward model as well as the wider retrieval framework not explored in Chapter 4. This chapter builds on the work of Warwick et al. (2022) by adapting the aircraft-based observations from this flight to mimic the expected FSI instrumental characteristics. As discussed in Chapter 1, Warwick et al. (2022) performed the first clear-sky retrieval on a single upwelling far- and mid-infrared spectrum observed during a science flight. These are referred to as ‘FORUM-aircraft’ observations and have been created following the approach described in Chapter 3. Section 5.2 describes the flight where the radiance observations were taken. Section 5.3 outlines which radiance observations were selected for the retrieval. Section 5.4 describes the optimisation of the IMS retrieval framework for retrievals from these FORUM-aircraft observations. The results from these retrievals are described in Section 5.5, and conclusions are drawn in Section 5.6.

5.2 Flight C153 Overview

The PIKNMIX-F campaign took place in March 2019, based out of Stornoway, Scotland and was a joint venture between the UK Met Office and Facility for Atmospheric Airborne Measurements (FAAM) Airborne Laboratory. The primary science goals were to obtain data that could be used to improve the representation of specific cloud-microphysical and boundary layer processes within the Met Office suite of models, and to validate radiative transfer modeling of cloudy scenes. Additional funding from ESA allowed further flight hours in support of the FORUM mission, such as the C153 flight that occurred on 13th March 2019.

The C153 flight path and altitude variation are shown in Figure 5.1. This flight aimed to observe simultaneous nadir-viewing spectra in the far- and mid-infrared using the TAFTS and ARIES instruments described in Chapter 3 (Canas et al. 1997, Wilson et al. 1999). Two straight and level runs (SLRs) were conducted between 57 and 55°N from 11:55 to 12:55 UTC

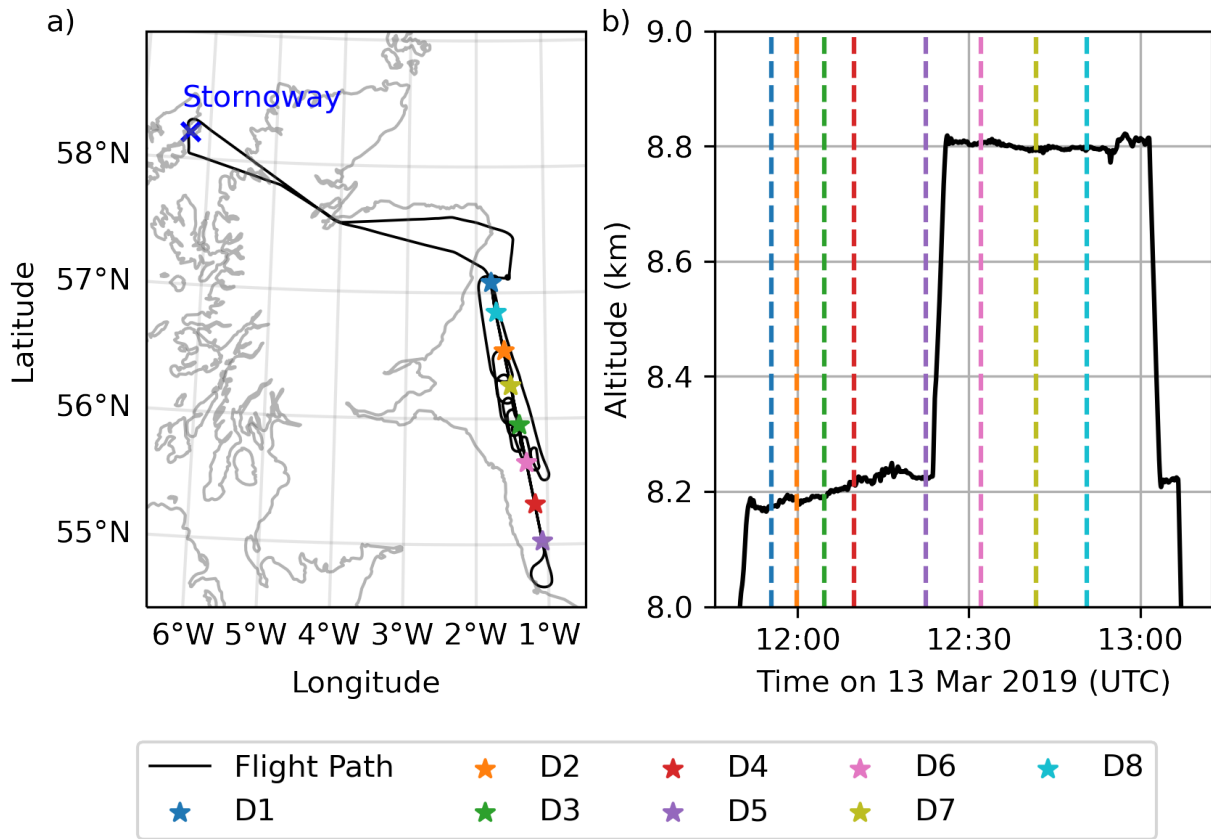


Figure 5.1: (a) The track of the C153 flight. (b) The altitude measured by the aircraft during the SLRs. Dropsonde releases are marked on (a) and correspond to the dashed lines in (b).

at speeds ranging from $175\text{--}190\text{ ms}^{-1}$. The first SLR travelled south-east at an altitude of ~ 8.2 km from the release of dropsonde 1 (D1) to the release of dropsonde 5 (D5). The aircraft then ascended and flew back over the same path at ~ 8.8 km, as highlighted in Figure 5.1b. After these SLRs were completed, the aircraft performed a spiral descent pattern followed by a final SLR at 300 m to the east of the main track, before it returned to Stornoway. Clear-sky conditions were generally observed throughout the flight, with a small amount of low cloud observed near the southern end.

5.2.1 Auxiliary Information

On board the FAAM aircraft, additional core instruments have been used to characterise the atmospheric state. Positional information was measured by the POS AV 410 GPS-aided Inertial Navigation unit, and static pressure was recorded by the Air Data Computer. Flight level ozone concentrations were recorded by a Core Thermo Fisher Scientific Inc. Model

49i UV absorption ozone photometer. Measurements from the aircraft's spiral descent after the second SLR have been used to build a vertical profile of ozone that is assumed constant throughout the flight and fixed in the retrievals.

The aircraft was also equipped with the Airborne Vertical Atmospheric Profiling System (AVAPS), which released eight Vaisala RD94 dropsondes throughout both SLRs (Figure 5.1b). Five dropsondes were released during the lower SLR and three during the higher SLR. The dropsonde measurements of water vapour are provided in relative humidity and mass mixing ratio. In this thesis, the concentrations of water vapour are expressed using the volume mixing ratio (units of ppmv). The conversions between these units are contained in Appendix B.

The Vaisala RD94 dropsondes use the same humidity sensor as the Vaisala RS92 radiosonde, whose uncertainty in humidity measurements was assessed in Miloshevich et al. (2009). The sensor calibration uncertainty was estimated as $\pm 5\%$ of the measured relative humidity value plus an absolute offset of $\pm 0.5\%$. The production variability uncertainty was $\pm 1.5\%$ of relative humidity values above 10% or $\pm 3\%$ for values below 10%. The procedure to convert these uncertainties into volume mixing ratio is described in Appendix B. No equivalent information is available for the temperature sensor, so a manufacturer quoted repeatability of 0.2 K is used to represent the uncertainty.

After release, each dropsonde drifted in a south-westerly direction away from the flight path, remaining within 25 km of their release point. The standard deviation of the ERA5 grid surrounding the path of each dropsonde is shown in Figure 5.2. Figure 5.2a suggests a low spatial variability of the water vapour between 4 and 6 km, and below 2 km. More variation in ERA5 water vapour is seen between 3 and 4 km, particularly around dropsondes 1 and 2, and above 6 km. Smaller variability is seen in the ERA5 temperature (Figure 5.2b) around the path of each dropsonde, however, the variation that is present peaks between 4 and 8 km and closer to the surface. Given the temporal and spatial resolution of the ERA5 data used (hourly and 0.25×0.25 degrees respectively), this variation can be taken as a lower limit on the in-situ variability that should be considered when evaluating the quality of the retrievals against the dropsonde profiles, and the variation between retrievals.

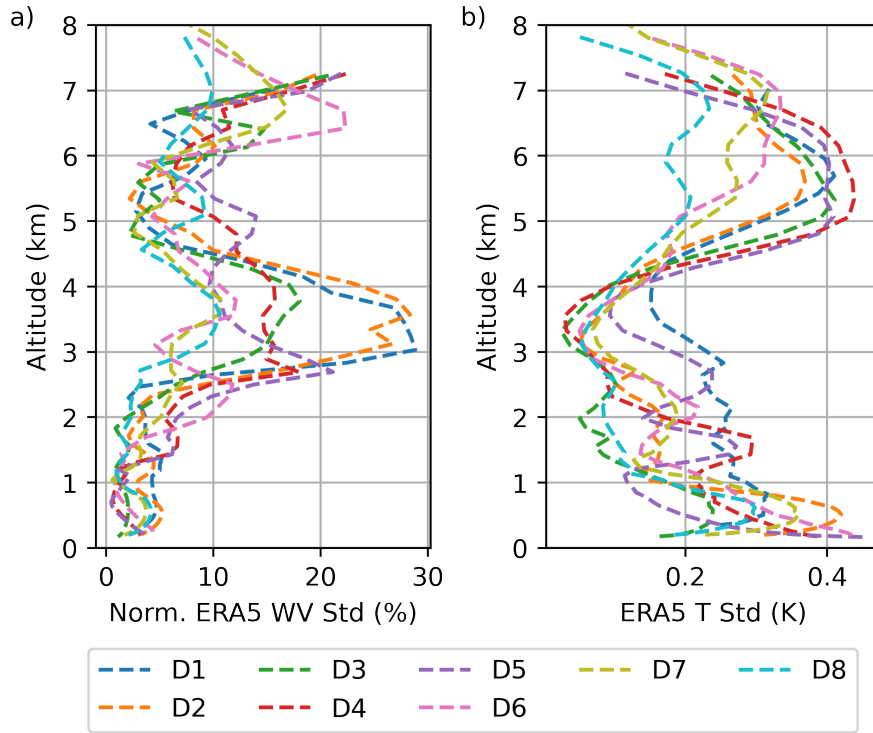


Figure 5.2: (a) The normalised standard deviation (%) of water vapour in ppmv (WV) and (b) standard deviation of temperature (T) taken from the ERA5 data surrounding the path of each dropsonde (± 30 mins, ± 48 km). The spatial resolution of ERA5 is $0.25 \times 0.25^\circ$ with a temporal resolution of one hour, and thus this plot gives an indication of the variability in the atmosphere at these scales.

5.3 Selection and Preparation for the Radiance Observations

The dropsondes provide an in-situ measurement of the atmospheric state. Therefore, the TAFTS observation that is closest in time to the release of each dropsonde has been selected. Six ARIES scans centred on each selected TAFTS spectrum were selected to create an average ARIES spectrum that is best matched to the TAFTS' field of view and scan time. This averaging reduces the random component of uncertainty however, the calibration uncertainty for ARIES tends to be relatively large between $600\text{--}1600\text{ cm}^{-1}$, and so averaging has only a small impact on the total instrument uncertainty (Figure 3.2b and c).

Final selection times are shown in Table 5.1. For dropsondes 3-5 and 8, there are observations from TAFTS and ARIES within 5 s of the dropsonde release. During the release of dropsonde 8, the ARIES instrument was calibrating and so there is a greater time difference between the TAFTS and ARIES measurements, however ARIES scans in this period are con-

Table 5.1: The times of each dropsonde release and the closest TAFTS scan in time from the C153 flight on 13 March 2019. For each TAFTS scan, the closest 6 ARIES scans have been averaged to provide the best match for the field of view and scan time between the instruments. The time of the central ARIES scan is shown.

Drops	Drops Release (UTC)	TAFTS (UTC)	ARIES (UTC)
1	11:55:29	11:57:17	11:57:17
2	11:59:53	11:59:43	11:59:42
3	12:04:43	12:04:47	12:04:47
4	12:09:55	12:09:58	12:09:58
5	12:22:27	12:22:23	12:22:22
6	12:32:12	12:32:07	12:32:07
7	12:41:49	12:40:43	12:40:46
8	12:50:39	12:50:39	12:51:07

sistent with simulations using the dropsonde 8 profile (Warwick et al. 2022). For dropsonde 2 the TAFTS and ARIES observations are 10 s away from the dropsonde release, and for dropsondes 1 and 7 there is over a minute difference which should be considered in later comparisons.

Following the method described in Chapter 3, these TAFTS and ARIES observations have been combined to create FORUM-aircraft observations, and are shown in Figure 5.3a-h with labels corresponding to their collocated dropsonde. These mimic the FSI instrument line shape and spectral sampling, and the gap in radiances between 300 and 330 cm^{-1} is a result of the gap between the TAFTS longwave and shortwave channels. As discussed in Chapter 3, this method of apodisation introduces an additional uncertainty in the 15 μm CO_2 band and between 1200-1400 cm^{-1} as a result of the coarse ARIES sampling. As a result, the uncertainty in the FORUM-aircraft observations is calculated by combining the instrument noise (Figure 3.2b), the instrument calibration uncertainty (Figure 3.2c), and the apodisation uncertainty (light blue in Figure 3.6) in quadrature. This is shown in red in Figure 5.3i.

The variability between all eight observations is shown in blue in Figure 5.3i in comparison to the instrument noise in yellow, and the FORUM-aircraft uncertainty in red. The variability between the eight spectra can be seen to exceed the noise throughout the FORUM-aircraft spectral range. This demonstrates a clear change of scene in the field of view between each

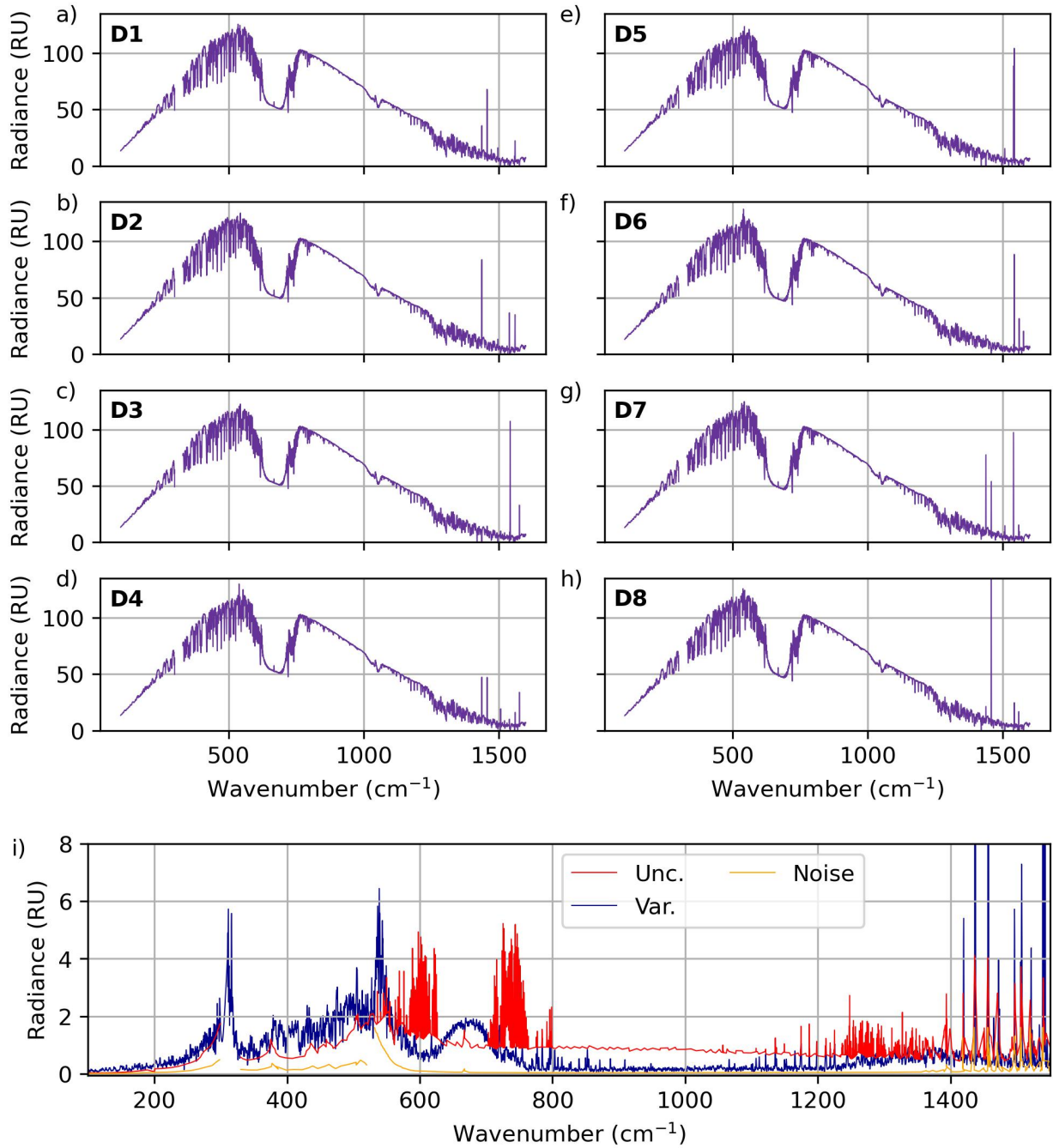


Figure 5.3: (a)-(h) The FORUM-aircraft observations selected in Table 5.1 labelled with their corresponding dropsonde (top left). Observed radiances are given in radiance units (RU) which are $\text{mW}/\text{m}^{-2}/\text{sr}/\text{cm}^{-1}$. Each FORUM-aircraft observation is composed of one TAFTS and six ARIES scans, as described in Chapter 3. (i) The uncertainty (Unc.) in each FORUM-aircraft observation is shown in red, and is the noise, calibration (Figure 3.2b and c) uncertainty, and apodisation uncertainty (light blue in Figure 3.6) combined in quadrature. The variability (Var.) across all eight FORUM-aircraft observations is shown in blue and represents one standard deviation. The noise (yellow) of a single FORUM-aircraft observation.

observation. This variability is largest near the detector band edges of TAFTS where the TAFTS calibration uncertainty increases, and in the large spikes seen above 1300 cm^{-1} in the ARIES noise and calibration uncertainty (Figure 3.2b and c).

To perform a retrieval on the selected FORUM-aircraft observations, the measurement covariance must be defined. As described in Section 2.2, the measurement covariance is used to constrain the retrieval and represents a combination of the uncertainty in the observation and the uncertainty associated with the forward model's calculations. Now that the uncertainty in the FORUM-aircraft observations has been evaluated, the next stage is to approximate the uncertainty in the forward model.

Specialised RTTOVv12 clear-sky coefficients have been developed by Richard Siddans (RAL Space), using LBLRTMv12.11, to enable simulations of upwelling spectra by RTTOVv12 at the altitude of each SLR with the spectral characteristics of the FSI. These coefficients were provided for use in this work, and their usage within IMS was performed by the author of this thesis. LBLRTMv12.11 uses the MT_CKDv3.5 continuum model, which, most notably for this study, contains an update to the water vapour continuum that increases water vapour absorption within the far-infrared (Mlawer et al. 2019).

The uncertainty associated with the RTTOVv12 simulations, and so the forward model, can be approximated by comparing the directly simulated radiances from LBLRTMv12.11 with RTTOVv12 simulations using the new coefficients. These residuals are shown in Figure 5.4 for both the TAFTS and ARIES spectral ranges. Typically, the generated differences were much smaller than the FORUM-aircraft uncertainty, excepting three or four isolated channels in the far-infrared (Figure 5.4a). These channels will be subsequently excluded from the channel selection process described in Section 5.4. Therefore, the uncertainty shown in red in Figure 5.3i is squared, and used as the main diagonal of the measurement covariance for the FORUM-aircraft configuration.

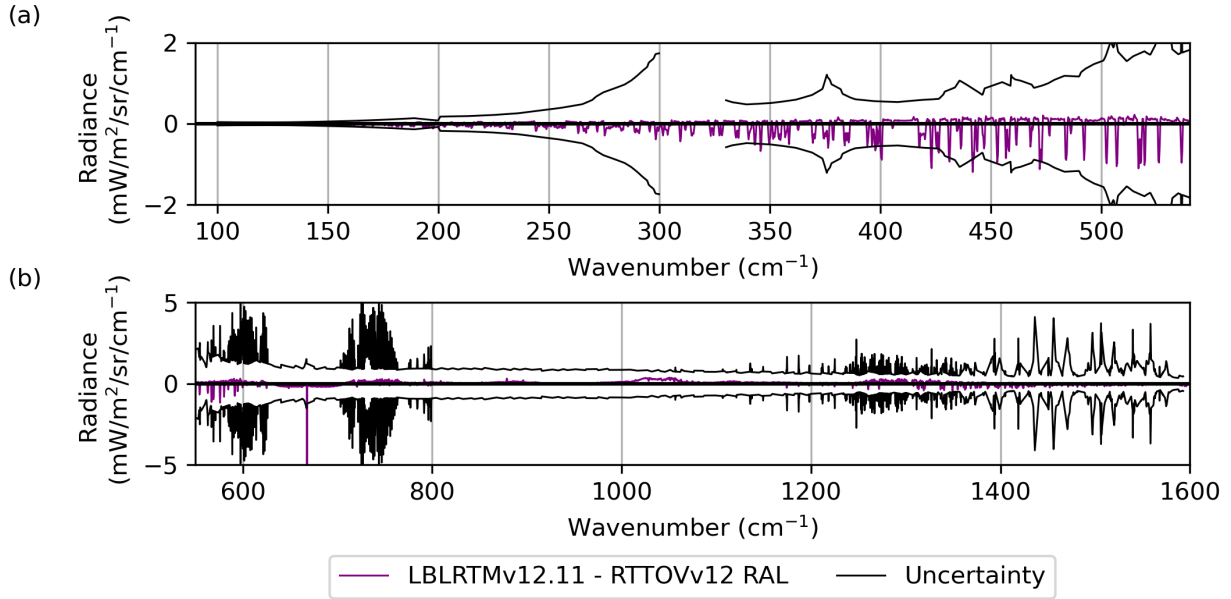


Figure 5.4: A summary of the uncertainties associated with the retrievals from FORUM-aircraft observations for the C153 flight. The purple line is the residual between an FSI apodised LBLRTMv12.11 simulation and RTTOVv12 FSI simulation using the aircraft-level coefficients in the (a) TAFTS and (b) ARIES spectral ranges. The dashed line is the same as in Figure 5.3i, and is the noise (Figure 3.2b) and calibration (Figure 3.2c) uncertainty, and apodisation uncertainty (light blue in Figure 3.6) combined in quadrature.

5.4 Channel Selection and Testing on Simulations

Using the FORUM-aircraft measurement covariance in Figure 5.5, new channels have been selected to optimise the information content available for the FORUM-aircraft configuration using the same approach as in Section 4.7.

The 200 channels with the highest information content are shown in Figure 5.5 and represent 63% of the total available information content, comparable to the FSI configuration's channel selection in Section 4.7. Similar to the FSI configuration, the FORUM-aircraft configuration selects channels in the 350-500 cm^{-1} region and the 15 μm (667 cm^{-1}) CO_2 band wings despite the larger measurement covariance (compare Figure 5.5 to Figure 4.10b). However, as the uncertainty for the FORUM-aircraft configuration is considerably reduced below 200 cm^{-1} relative to the rest of its spectral range, 23% of its channels have been selected in this region while none were identified for the FSI configuration in this range.

To compare the performance of the FORUM-aircraft against the FSI configuration, the testing in Section 4.7 has been repeated using the newly selected channels and measurement

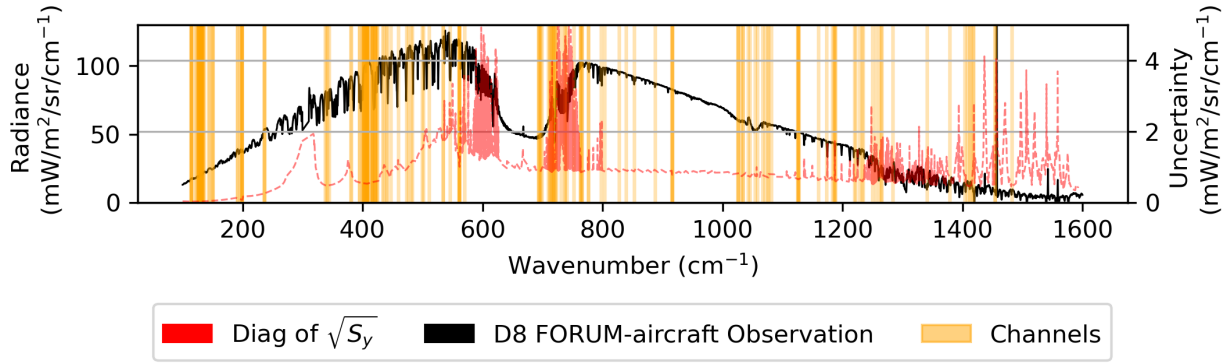


Figure 5.5: The square-root-diagonal of the measurement covariance (equivalent to the FORUM-aircraft uncertainty (Figure 5.3i) overlaid on the FORUM-aircraft observation closest to dropsonde 8 (Figure 5.3h). This uncertainty is composed of the instrument uncertainty and the uncertainty that arises from the FORUM-aircraft apodisation process discussed in Section 3.3. The instrument uncertainty is the radiometric calibration and random noise for each instrument combined in quadrature for a single TAFTS scan and six ARIES scans. For both instruments, this uncertainty is dominated by the calibration, particularly for ARIES, where the random noise is reduced due to multiple scans (Figure 3.2b and c). The impact of the FORUM-aircraft apodisation process can be seen most clearly in the $15 \mu\text{m}$ CO_2 band and between $1200\text{--}1400 \text{ cm}^{-1}$. The 200 channels selected for the aircraft retrievals are also shown in orange.

covariance calculated for the FORUM-aircraft configuration. These FORUM-aircraft retrievals have been performed using the same test cases outlined in Section 4.4 but for an observing sensor at the pressure level of the higher SLR of the C153 flight. Figure 5.6 compares the temperature and water vapour bias between the FSI and FORUM-aircraft configurations. Generally, there is an improvement with the FORUM-aircraft configuration of up to 2 % and 0.1 K in the water vapour and temperature bias, respectively, against the FSI configuration below the pressure level of the higher SLR. However, the ESD associated with the FORUM-aircraft configuration is up to 15 % and 0.6 K larger for water vapour and temperature, respectively, in the mid- to lower troposphere. The median DOFS for temperature has also reduced by ~ 0.4 due to the larger measurement covariance in the CO_2 band wings.

Figure 5.7 shows the median surface emissivity and skin temperature retrieved from the 240 test simulations using both of the configurations (FSI and FORUM-aircraft) created within this thesis. Some minor improvements are observed with the FORUM-aircraft configuration. Due to the larger number of channels below 250 cm^{-1} for the FORUM-aircraft configuration, the median emissivity residual (Figure 5.7a) only reaches a maximum of 0.014 in this spectral

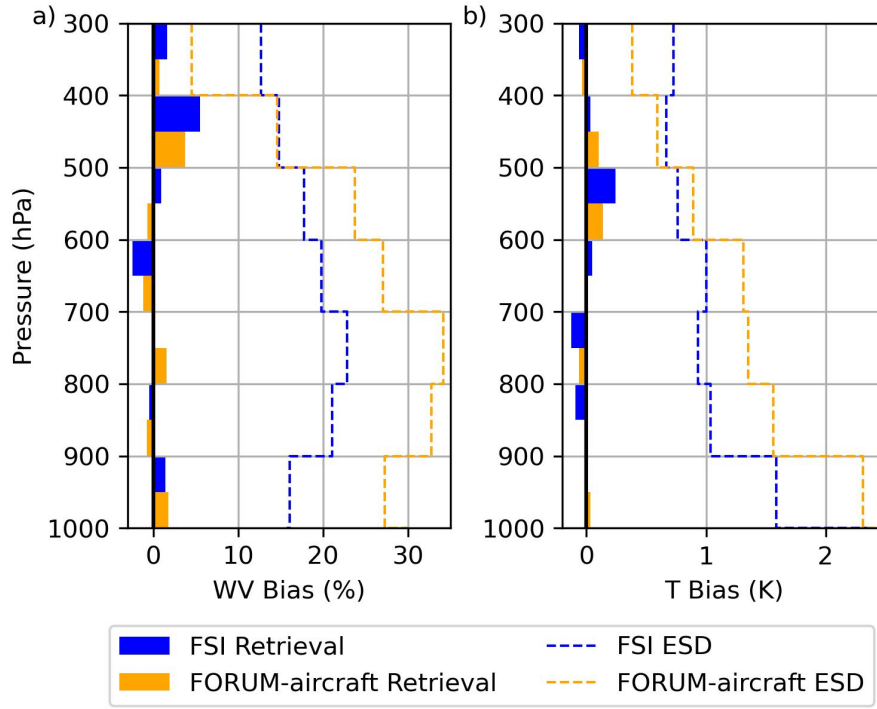


Figure 5.6: The median (a) water vapour (WV) and (b) temperature (T) bias of the retrieved profile to the true state across the testing cases for the optimised FSI and FORUM-aircraft configurations in blue and orange respectively. Biases are evaluated between 300 and 1000 hPa in 100 hPa bins as in Trent et al. (2023). Dashed lines show the median estimated standard deviation (ESD) for a single retrieval for the test cases in each bin. For water vapour this is plotted with an offset of - 10% for clarity.

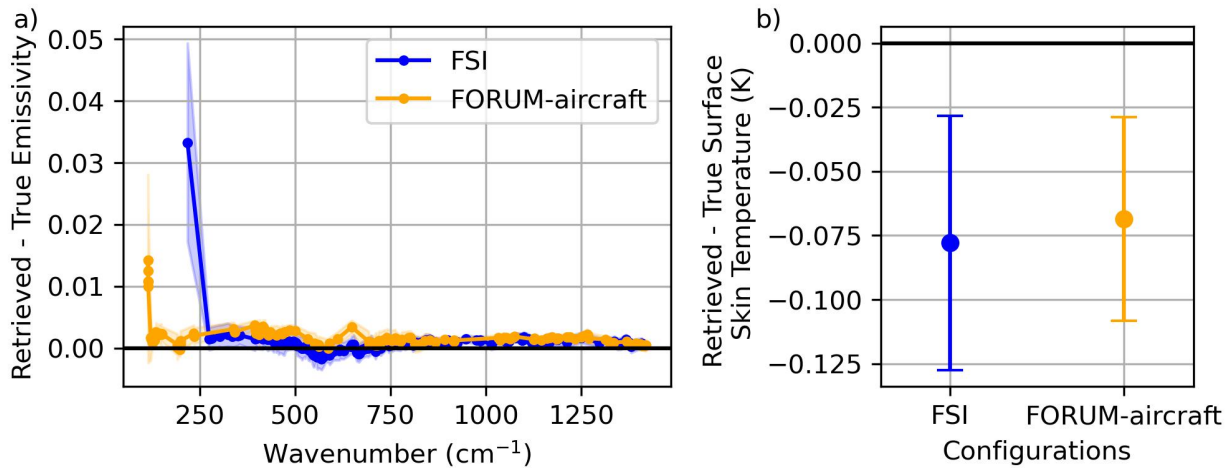


Figure 5.7: The median residual between the retrieved and input (a) surface emissivity and (b) surface skin temperature for the FSI and FORUM-aircraft configurations across the 240 cases in blue and orange, respectively. The shaded regions and error bars represent the median absolute deviation around this residual. The FSI emissivity residual shown here is the same as in Figure 4.14.

region as it is more tightly constrained. In addition, there is also a reduction of 0.01 in the median absolute deviation of the retrieved skin temperature across all 240 cases (Figure 5.7b). However, both configurations retrieve the emissivity within 0.01 of the ‘true’ value above 250 cm^{-1} , and have a comparable median retrieval bias for the skin temperature within ~ 0.01 K of each other.

The similarity between the FORUM-aircraft and FSI retrieval performance across the test cases hints that when the FORUM-aircraft configuration is applied to real observations in the next section, the results will give a realistic indication of the potential quality of retrievals that can be expected from the FSI.

5.5 Retrievals from Aircraft Observations

5.5.1 Initial Retrieval

Retrievals of the surface skin temperature, spectral surface emissivity, temperature, and water vapour were performed on the aircraft observations selected in Section 5.3 that were modified to mimic the FSI’s instrument line shape. In each case, the closest hourly ERA5 data was used as the a-priori for surface skin temperature, atmospheric temperature, and water vapour, accompanied by the weak, global a-priori covariance outlined in Section 4.2.

The average retrieved surface skin temperature was found to be 280.3 ± 0.2 K across the eight cases. This is consistent with findings from the iterative method used in Warwick et al. (2022) to derive the surface skin temperature and agrees with radiance observations from the 300 m aircraft run. Similarly, Figure 5.8 shows that all of the eight retrieved surface emissivities agree within one ESD. Below 120 cm^{-1} , the ESD for surface emissivity in all eight cases increases up to 0.08 due to the limited sensitivity to surface emissivity at lower wavenumbers, as observed in the simulated test cases in Section 5.4.

Figure 5.9 shows the mean temperature and water vapour retrievals from all eight observations (red curves) collocated with the release of each dropsonde. The black curve shows the average of all the dropsonde measurements that have had their respective averaging kernel applied using the theory in Section 2.2.3 and Eq. 2.43. In this thesis, these are referred

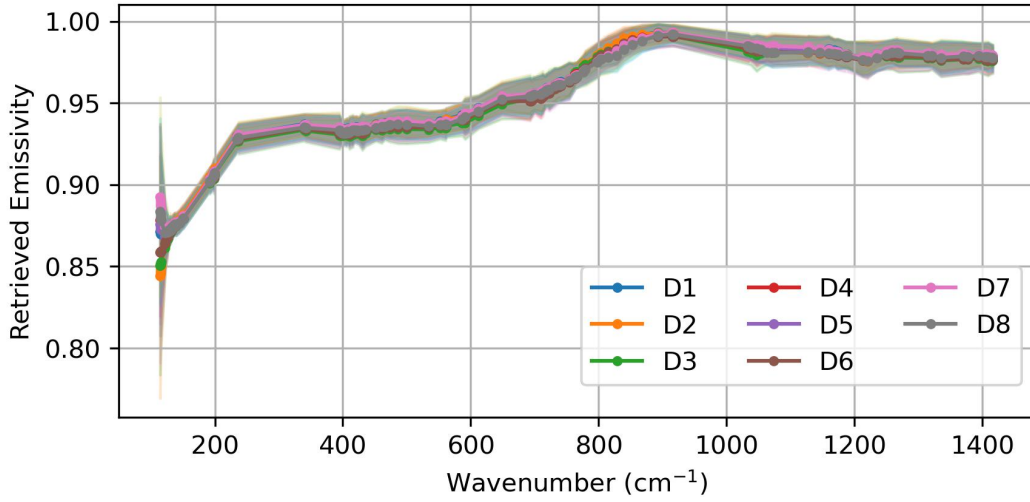


Figure 5.8: The retrieved surface emissivity in each of the 200 channels used in the retrievals from the eight spectra selected in Section 5.3. The shading represents one ESD in the retrieved value. The mean retrieved surface skin temperature was found to be 280.3 ± 0.2 K.

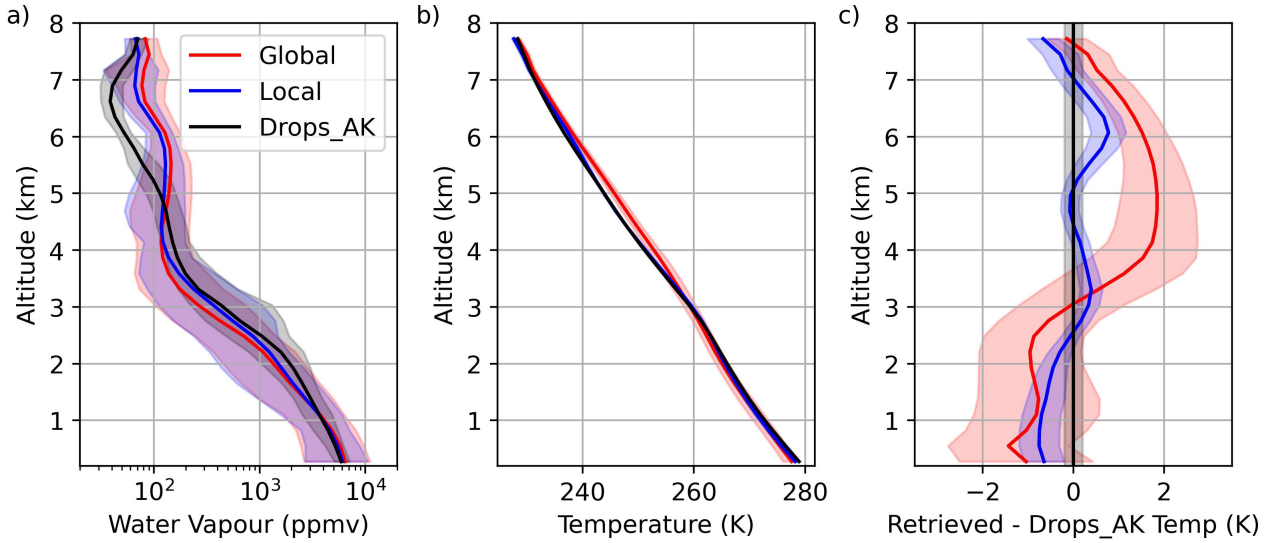


Figure 5.9: The mean retrieved (a) water vapour and (b) temperature profiles, as well as the (c) residual between the retrieved temperature and each AK-treated dropsonde profile (Drops_AK). Retrievals are shown using the global and local grid a-priori covariances for temperature in red and blue, respectively. The Drops_AK water profile and temperature profiles shown in (a) and (b) did not change significantly with the different covariances and so are shown only for the global covariance. The temperature residuals shown in (c) were calculated with respect to the correct Drops_AK profile. The global a-priori covariance is calculated from ERA5 temperatures across the globe for three days as used in Trent et al. (2023). The local a-priori covariance is calculated from ERA5 temperatures surrounding the SLRs for the duration of the flight. Blue and red shaded regions show the full spread of retrieved profiles with purple regions showing the overlap. The grey shaded region in (a) shows the full spread of AK-treated dropsonde profiles as well as the measurement uncertainty, and in (b) and (c) shows the dropsonde measurement uncertainty only.

to as the AK-treated dropsonde profiles. The water vapour retrieval captures the AK-treated dropsonde profile below approximately 5 km within its uncertainty. However, the temperature retrieval has a limited performance, particularly above 4 km and near the surface, with a mean retrieval bias of up to 2 K.

Warwick et al. (2022) indicated the ARIES spectra were, at best, poorly constrained in regions of the spectrum sensitive to atmospheric temperature due to possible heating of the instrument housing. Here, the additional impact of the apodisation in the CO₂ band on the measurement exacerbates this unreliable information and results in the large temperature residuals between the AK-treated dropsonde and retrieved profiles as the retrieval attempts to fit anomalously warm observations. It is unlikely this is due to the dropsondes' representation of the state. While Figure 5.2b shows a variation of up to 0.5 K in ERA5 around each of the dropsondes between 4 and 7 km, a systematic effect is observed across all of the retrievals.

To compensate for this, the a-priori covariance for temperature has been tightened to what will be referred to as the local covariance, and is shown in Figure 5.10. This local covariance was calculated by directly calculating the covariance of ERA5 temperature profiles

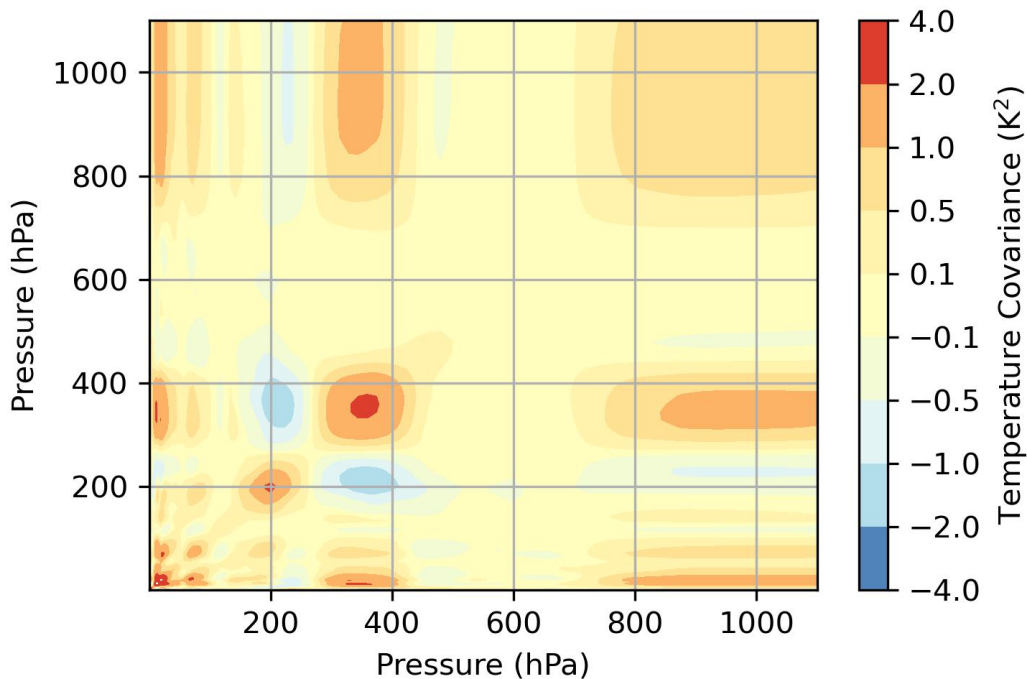


Figure 5.10: The local a-priori covariance for temperature used in retrievals from aircraft observations. This was calculated using ERA5 data surrounding the SLR for the duration of the flight (54.5-57.25°N and 6.25-1.75°W for 11:00-13:00 UTC).

surrounding the SLRs for the duration of the flight. The a-priori covariance for water vapour has not been tightened and is still derived from global ERA5 data across three days. The impact on the mean temperature and water vapour AKs across the 8 cases is shown in Figure 5.11. As expected, Figure 5.11d shows a reduction in the temperature AKs, with a 60% reduction in the average temperature DOFS. This tightening of the a-priori temperature covariance also leads to an increase in the water vapour AKs focused below 4 km with a 17% increase in the water vapour DOFS in Figure 5.11b.

Figure 5.9 shows that the implementation of the local temperature constraint significantly improves agreement between the temperature retrievals (blue curve) and the AK-treated dropsonde profiles, with a mean temperature residual of at most 0.8 K. A similar improvement is observed in the percentage difference between the water vapour retrievals and AK-treated dropsonde, with an improvement of up to 20 % at 3 km, and up to 33 % at 6 km. A smaller effect is observed in the retrievals at higher altitudes. Only a negligible impact is observed on the retrieved surface properties. This method enables the joint retrieval of temperature, water vapour, surface emissivity, and surface skin temperature that was not possible in Warwick et al. (2022) due to the outlined limitations for the ARIES spectra.

5.5.2 Revised Retrievals

Given the findings in Section 5.5.1, this section analyses the retrievals performed using the local a-priori covariance for temperature derived from ERA5 temperature profiles surrounding the SLRs for the duration of the flight, and the original global a-priori covariance for skin temperature and water vapour. The surface emissivity a-priori covariance defined in Chapter 4 is still used, and was derived from the IREMIS atlas for sea surface types, and using the Huang et al. (2016) emissivity atlas for land surface types, as outlined in Section 4.6.2.

Figure 5.12 shows each retrieved water vapour and temperature profile compared to the a-priori, and the reference AK-treated and untreated dropsonde profiles. The retrieval typically brings the water vapour profile from the a-priori estimate towards the AK-treated dropsonde profile. This is reflected by an 18% reduction in the Root-Mean-Squared-Error (RMSE) relative to AK-treated dropsonde profiles throughout the vertical profile (Table 5.2).

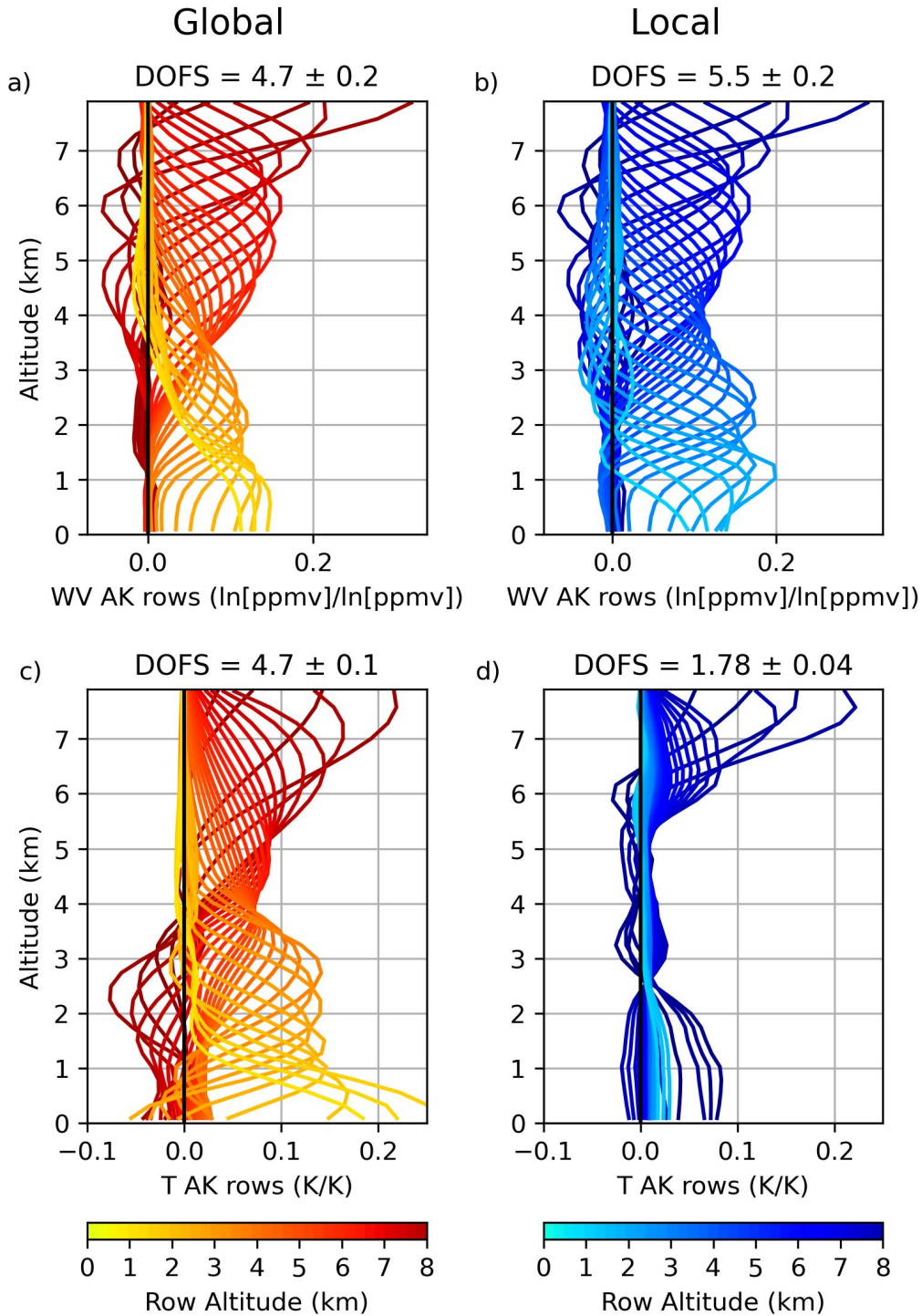


Figure 5.11: Rows of the mean averaging kernels (AK) for (a,b) water vapour and (c,d) temperature calculated from retrievals of the C153 observations with colours representing the dropsonde altitude of each profile level. (a) and (c) follow the same colour scheme and are the rows of the AKs when the a-priori temperature covariance was derived from three days of global ERA5. (b) and (d) follow the same colour scheme and are the rows of the AKs when the a-priori temperature covariance was derived from the ERA5 surrounding the SLRs for the duration of the flight. The water vapour a-priori covariance used is the same in both sets of retrievals. The mean and standard deviation of the respective DOFS from the eight retrievals are shown for each plot.

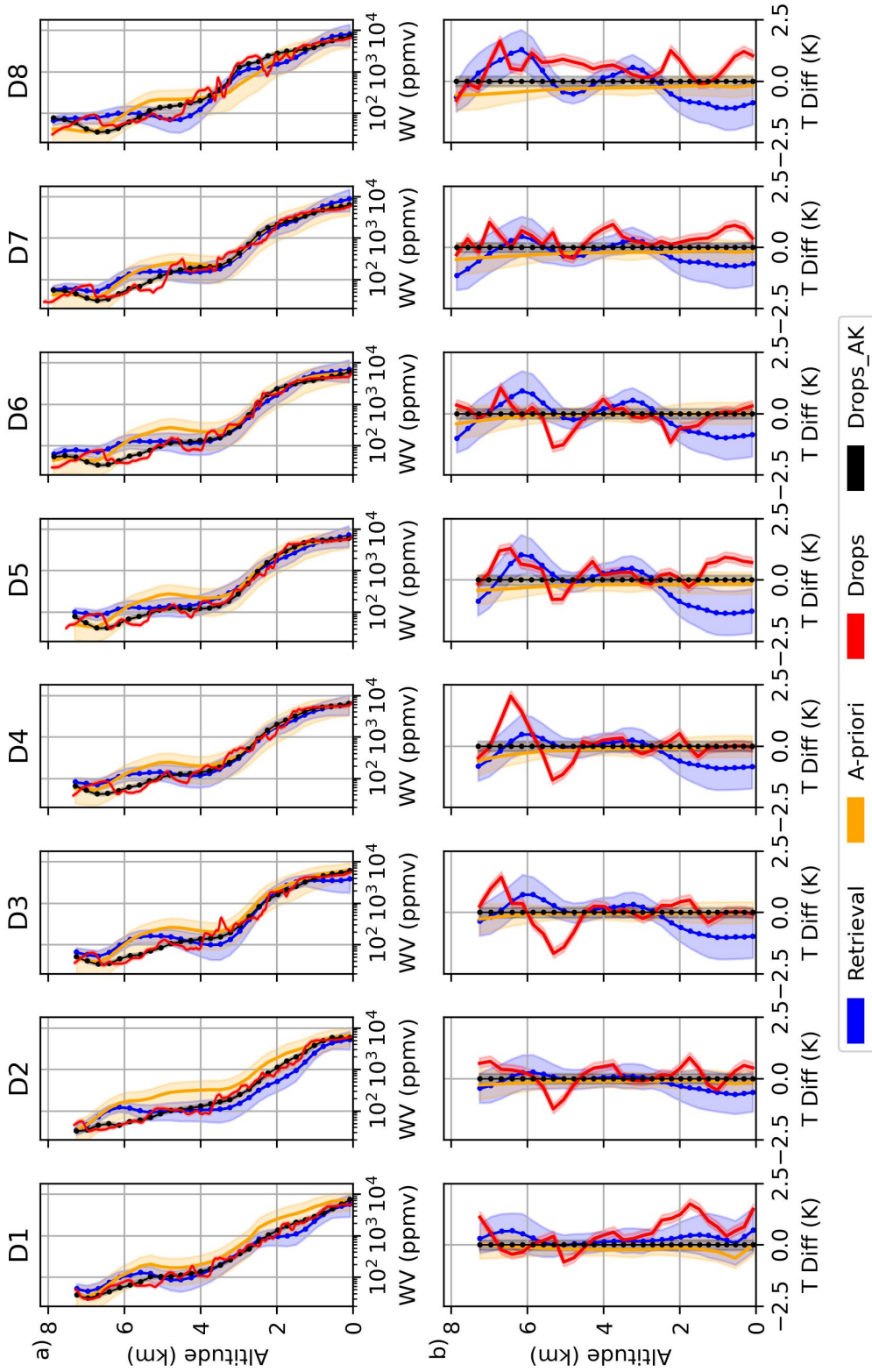


Figure 5.12: (a) Water vapour (WV) retrievals and (b) the difference between the retrieved temperature and AK-treated dropsonde profiles (T Diff) performed on the FORUM-aircraft radiances. The shaded region in each retrieved value represents two ESD of the retrieval derived from the square-root diagonal of S_x . The a-priori is shown in orange, with the shaded region showing the a-priori standard deviation. The dropsonde profiles from each case are shown in red, and each one has had their respective averaging kernels applied (Drops_AK), which are shown in black. The shading around the dropsonde and Drops_AK profiles represents the uncertainties outlined in Section 5.2.1. For temperature, the dropsonde uncertainty is represented as 0.2 K. For water vapour, the dropsonde uncertainty varies with altitude but remains below 9%.

Table 5.2: The RMSE value relative to each AK-treated dropsonde profile for temperature and water vapour for the 8 observations against the a-priori (ERA5) and retrieved profiles. To aid later interpretation, RMSE values have been separated into above and below 5 km, alongside those for the full profile.

	A-priori	Retrieval
Full T (K)	0.21	0.57
Full WV (log(kg/kg))	0.50	0.41
Below 5 km T (K)	0.15	0.59
Above 5 km T (K)	0.29	0.53
Below 5 km WV (log(kg/kg))	0.41	0.30
Above 5 km WV (log(kg/kg))	0.65	0.57

Retrievals of temperature have a restricted movement from the a-priori as outlined in Section 5.5.1, and as a result, applying the AKs in Figure 5.11d brings each dropsonde profile to within 0.6 K of their corresponding a-priori.

Both the retrievals of temperature and water vapour generally capture their reference state within their uncertainty. However, some issues are apparent: for example, the water vapour retrievals struggle with a persistently dry layer measured by the dropsondes between 5 and 7 km; and the temperature retrievals generally have a warm bias above 5 km, and a cold bias closer to the surface. A deeper analysis of the retrievals follows, divided into altitudes above and below 5 km. To aid this analysis, Figure 5.13 shows (a) the average weighting function for selected 100 cm^{-1} wavenumber bands, (b) the spectral residuals between the final retrieved state and the observations, and (c) the improvement in the radiance residuals from those simulated using the a-priori estimate.

Above 5 km

Figure 5.12a shows that the retrieved water vapour concentration is generally overestimated relative to the dropsonde measurements at altitudes above 5 km. The most obvious discrepancy occurs in a layer between approximately 5 and 7 km, where the dropsondes are persistently drier than the a-priori estimate. Although, in most cases, the retrieved profiles (blue lines) move towards the corresponding AK-treated dropsonde profiles (black lines) between 5 and 6 km, the shift is small and the difference between them can exceed 2 ESDs of

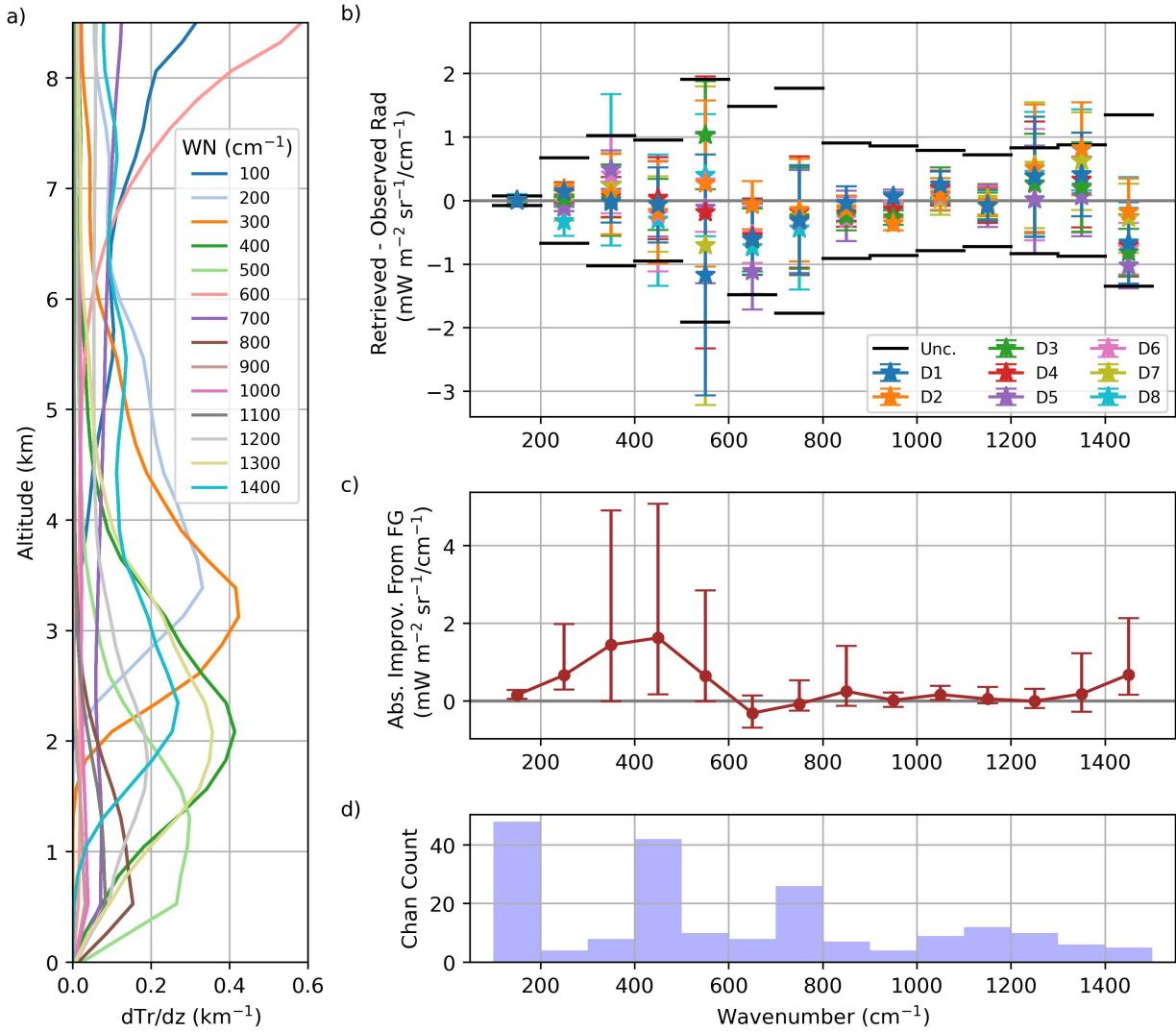


Figure 5.13: (a) The mean weighting function (change in transmission (Tr) with altitude (z)) for each wavenumber (WN) bin averaged across the 200 channels for all 8 dropsondes. (b) The radiance residual between the retrieved and FORUM-aircraft observed spectra for all 8 cases in each bin. Residuals have been averaged over 100 cm⁻¹ bins, and the mean and standard deviation are shown for each bin. The black line represents the mean uncertainty (Unc.) from Figure 5.5, which is the square-root diagonal of S_y , in that bin. (c) The mean absolute improvement in the spectral residual from the first guess (FG) spectra to the retrieved spectra across all 8 cases in each bin. Error bars show the minimum and maximum values across the 8 cases in each bin. (d) The number of channels in each bin.

the retrieval. Above 6 km, movement from the a-priori towards the AK-treated dropsonde is less obvious.

As the AK-treated dropsonde also does not deviate substantially from the original dropsonde profile in this region, it is not expected that these differences are caused by a weak sensitivity to the profile or by the a-priori. Figure 5.13a shows the channels with the highest

sensitivity between 5 and 7 km tend to be clustered between 100 and 400 cm^{-1} and above 1400 cm^{-1} , within the water vapour rotation and vibration-rotation bands. Between 100-400 cm^{-1} water vapour spectroscopy, including line and continuum contributions, has been continuously updated over the last couple of decades but is still subject to relatively large uncertainty (Mlawer et al. 2019, 2023), which will contribute to forward model error. Above 1400 cm^{-1} the fitted radiances are on average underestimated by $-0.6 \text{ mWm}^{-2}\text{sr}^{-1}/\text{cm}^{-1}$, however this remains within the measurement covariance (Figure 5.13b). Recent work in Fox et al. (2024) found that the RD94 dropsondes used in FAAM flights between 2015 and 2021 tended to have a dry bias in comparison to the newer models as a result of contamination during long storage times. This could also contribute to the observed differences between the retrieved and measured water vapour profiles. However, specific information about this dry bias was not provided.

The temperature retrievals broadly capture the AK-treated dropsonde within the uncertainty of the retrieval but generally do not present an improvement from the a-priori. As discussed in Section 5.5.1, an overestimation of up to 1.3 K is seen above 5 km due to the ARIES spectra. However, there is also an underestimation observed closer to the aircraft. In Warwick et al. (2022), there were notable differences between simulated radiances and ARIES observations between 650-700 cm^{-1} . Six out of the eight channels in the 600-700 cm^{-1} bin are within this spectral range. As Figure 5.13a shows, the channels used within this bin show a greater sensitivity to the atmosphere above 7 km, and the fitted radiances within this bin are generally underestimated (Figure 5.13b).

Below 5 km

When making comparisons to the dropsonde profiles at lower altitudes, it is important to note that the dropsondes have travelled up to 23 km from their release when they reach the surface. This is most significant for the retrievals closest to dropsonde 1 and 7 where the radiometric observations are taken over a minute from the dropsonde releases and so have a weaker reference of the atmospheric state at lower altitudes. Furthermore, the ERA5 reanalyses surrounding each dropsonde in Figure 5.2 imply a higher variability in the water vapour

concentrations between 2 and 4 km, reaching up to 30 % in water vapour for dropsonde 2. The corresponding temperature standard deviation reaches up to 0.5 K closer to the surface for all dropsondes. Hence it is feasible that the dropsonde profiles are less representative of the atmosphere sounded by the aircraft spectrometers at these altitudes.

Nonetheless, below 5 km the a-priori for water vapour for dropsondes 3-7 is close to the dropsonde profile, particularly below 2 km. For dropsondes 1, 2, and 8, there is a larger discrepancy that the retrieval successfully reduces, shifting the retrieved values closer to the relevant AK-treated dropsonde profile. Indeed, in all cases except dropsonde 2, the retrieval uncertainty fully encompasses the dropsonde values below 4 km. Even for the case closest to dropsonde 2, the retrieval significantly shifts from the a-priori closer to the dropsonde profile, with the largest spectral shifts observed between 200 and 600 cm^{-1} (Figure 5.13b).

Between 2 and 5 km, all the temperature retrievals capture the AK-treated dropsonde profile within their retrieval uncertainty with limited movement from the a-priori as would be expected from the AKs (Figure 5.11d). Nearer the surface, the average retrieval bias for the retrievals closest to dropsondes 2-8 is -0.9 ± 0.2 K relative to the AK-treated dropsonde profiles. Of these cases only the retrieval closest to dropsonde 5 underestimates the AK-treated dropsonde temperature outside of both their uncertainties, and this by only 0.1 K. The retrieval closest to dropsonde 1 is the only case that shows a warm bias to the AK-treated dropsonde profile nearer the surface, however as previously mentioned, in this instance the dropsonde provides a weaker reference of the true profile.

These temperature and humidity changes are manifested in radiance space as shown in Figure 5.13b. Figure 5.13a highlights that the spectral regions between 400 and 600 cm^{-1} and 1200 and 1400 cm^{-1} are most sensitive to lower altitudes, particularly below 3 km. In these bins, particularly in the far-infrared, the retrieval reduces the spectral residuals by up to 5 $\text{mWm}^{-2}\text{sr}^{-1}/\text{cm}^{-1}$ from the first guess spectra (Figure 5.13c). Despite these improvements, the retrievals closest to dropsondes 1-3 still demonstrate a limited fit in the 500-600 cm^{-1} and 1300-1400 cm^{-1} bins. While the timing of dropsonde 1 is somewhat offset from the radiometric aircraft measurements the same is not true for dropsondes 2 and 3. Dropsondes 1 and 2 also show enhanced water vapour variability between 3 and 4 km in the surrounding

ERA5 profiles compared to the other cases (Figure 5.2b), but this enhancement is not seen around dropsonde 3.

5.6 Conclusions

This chapter presents the first application of the extended Infrared Microwave Sounding retrieval scheme to far- and mid-infrared radiances observed in clear-sky conditions. Retrievals of temperature, water vapour, surface emissivity, and surface skin temperature have been performed on observations of upwelling radiances measured from the upper troposphere and compared to in-situ measurements of the atmospheric state.

Coincident aircraft observations of far- and mid-infrared radiances were used to test IMS on clear-sky cases. These were taken during an aircraft flight in the upper troposphere, with periodic release of dropsondes to measure the atmospheric state. Eight observations closest to each dropsonde release were selected and modified to emulate the FORUM Sounding Instrument's spectral characteristics following the method described in Chapter 3. The combination of the aircraft and FSI spectral characteristics resulted in an increased spectral uncertainty, particularly in the $15\text{ }\mu\text{m}$ CO_2 absorption band. This propagated through to change the channels selected for the retrievals when compared to the top-of-atmosphere simulation study. Applying this revised 'FORUM-aircraft' configuration to simulated aircraft level radiances from the MERRA-2 test set showed only a limited impact on the retrieved temperature and water vapour profiles, compared to values derived using the original FSI configuration from the equivalent TOA radiances. This gives confidence that the results obtained here from the aircraft observations are indicative of what might be expected from the FSI when it is operating.

Retrievals of the FORUM-aircraft observations required the a-priori covariance for temperature to be tightened to the local ERA5 grid of the SLRs for the duration of the flight due to the larger measurement uncertainty associated with the observed spectra, particularly across the $15\text{ }\mu\text{m}$ CO_2 band. This enabled simultaneous retrievals of temperature and water vapour, exploiting the full FSI spectral range. Tightening this constraint had the additional

benefit of reducing the bias in the water vapour retrieval by up to a third. The final retrievals of temperature and water vapour generally captured the dropsonde measurements of the atmospheric state, with the RMSE in water vapour concentration reducing by 18 % from the a-priori to the retrieved state across all eight cases. These results do still highlight some inconsistencies, with both the ERA5 a-priori and retrievals consistently overestimating the humidity between 5 and 7 km, and a persistent cold bias below 2 km in seven out of the eight cases analysed. These deficiencies are manifested in the radiance residuals seen in the final, fitted spectra and suggesting that the humidity bias may be related to water vapour spectroscopic uncertainty in the far-infrared. However, these differences may also be caused by a dry bias observed in the model of dropsondes used in recent work. While this analysis implies that the variability around each dropsonde is relatively small, the near surface bias may be partly related to the dropsonde drift away from the aircraft as it descends through the atmosphere.

To summarise, the Infrared Microwave Sounding scheme has now been tested on multiple aircraft-level observations of upwelling far-infrared radiances, in addition to a diverse set of top-of-atmosphere simulated upwelling radiances, exploiting channels from the full FSI spectral range. Pending the real FSI systematic and random uncertainties, IMS is ready to use for clear-sky retrievals from the FSI.

Chapter 6

First Retrievals of Ice Cloud Properties from Far-Infrared Observations

Key Points

- This chapter contains the first retrieval of ice cloud properties (cloud effective radius, cloud top height, cloud optical thickness, ice crystal habit) from an upwelling observation of far- and mid-infrared radiances taken at aircraft level and evaluates it against in-situ measurements of the cloud.
- 26 channels have been selected based on their sensitivity to optimise the retrieval of ice crystal properties for this case study. Additional coefficients have also been built to enable retrievals of ice crystal habit.
- Tests showed that the approximations used within this retrieval framework to simulate and retrieve from cloudy radiances do not compromise its ability to retrieve the 'true' state in this case.
- The inclusion of the far-infrared halved uncertainties in the retrieved cloud top height and optical thickness, and enabled a distinction between ice crystal habits which is in line with previous findings. However, cloud optical property models still show limitations in the far-infrared.

6.1 Introduction

The IMS retrieval framework has now been applied to observations of upwelling far- and mid-infrared clear-sky radiances that have been modified to mimic the expected FSI spectral characteristics. The next stage of this thesis is to apply IMS to upwelling radiances that have been observed from above a cirrus cloud to investigate the significance of including the far-infrared.

The approximations used to retrieve cloud properties by IMS prior to this thesis were built by Richard Siddans (RAL Space) and are described in Section 6.2. The remainder of this chapter is original work produced independently by the author of this thesis unless specified otherwise in the text. Section 6.3 then introduces the flight where the radiance observation and in-situ measurements of the cloud were taken. Section 6.4 details the changes that have been made to IMS to optimise the retrieval for this case study. The limitations of IMS are then examined in Section 6.5 through retrievals of visible cloud optical thickness (COT), cloud effective radius (CER), cloud top height (CTH), and ice crystal habit using a fixed temperature and gaseous profile from the observation and from a comparable simulation. Simultaneous retrievals of temperature, water vapour, visible COT, CER, CTH, and ice crystal habit are then performed on the observation taken during a campaign flight in Section 6.6, and evaluated against in-situ measurements of the cloud and atmospheric state. Conclusions are drawn in Section 6.7.

6.2 Approximations Used to Retrieve Ice Cloud Properties

IMS can currently retrieve the visible COT, CER, and CTH simultaneously with the temperature and water vapour profiles. Building on the initial description of IMS and RTTOVv12 in Section 4.2, this section details the existing approximations used in RTTOVv12 (Section 6.2.1), and the wider retrieval framework (Section 6.2.2) to perform retrievals of ice cloud properties.

6.2.1 Simulating the Radiances

RTTOVv12 uses the Chou approximation to simulate cloudy radiances as it increases operational speed. This approximation scales the optical depth associated with absorption and emission to include the effects of scattering and then calculates the upwelling radiance by substituting this optical depth into the clear-sky radiative transfer equation (Eq. 2.9). A brief outline of the approximation is provided here and is taken from Chou et al. (1999) and Matricardi (2005).

To derive the Chou approximation, the contribution of a layer to the upward radiation at τ^* is considered, shown in Figure 6.1. The apparent emission, $dI'(\tau, \mu)$, of a layer with thickness $d\tau$ can be derived from the radiative transfer equation for upwelling radiation in Eq. 2.19:

$$dI'(\tau, \mu) = (1 - \omega_0)B[T(\tau)] \quad \text{Atmospheric Absorption and Emission} \\ + \frac{\omega_0}{2} \int_{-1}^1 I(\tau, \mu_i) \overline{P}(\mu, \mu_i) d\mu_i \quad \text{Scattering} \quad (6.1)$$

where the first term describes the atmospheric emission of the layer, and the second term

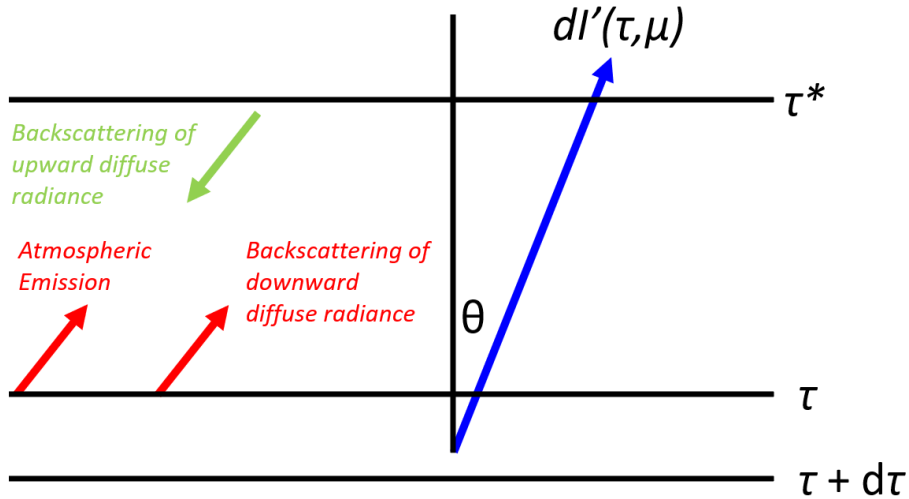


Figure 6.1: A schematic of the contribution of a layer to the upward radiance observed at an altitude where $\tau = \tau^*$. This is adapted from Matricardi (2005), and excludes the scattering of solar radiance.

corresponds to the scattered radiation that has been backward scattered from above or below the layer. The variables used within this equation were introduced in Section 2.1, and are summarised in Table A1 in the Appendix.

The Chou approximation assumes that the radiance field, $I(\tau, \mu_i)$, is diffuse and isotropic, and that it can be approximated using a Planck function ($I(\tau, \mu_i) \approx B[T(\tau)]$). This means that the second term on the right-hand side of Eq. 6.1 can be rewritten as:

$$\frac{\omega_0}{2} \int_{-1}^1 I(\tau, \mu_i) \bar{P}(\mu, \mu_i) d\mu_i = \omega_0 b B[T(\tau)] \frac{d\tau}{\mu} \quad (6.2)$$

where b is the backscattering parameter which represents the fraction of incident radiance (from above or below) that is scattered backwards, and is defined using Eq. 6.3:

$$b = \left[\frac{1}{2} \int_0^1 d\mu \int_{-1}^0 \bar{P}(\mu, \mu_i) d\mu_i \right]. \quad (6.3)$$

By substituting Eq. 6.2 into Eq. 6.1, the absorption and emission and scattering terms on the right-hand side of Eq. 6.1 can be combined:

$$dI'(\tau, \mu) = [1 - \omega_0(1 - b)] B[T(\tau)] d\tau / \mu \quad (6.4)$$

and used to define the apparent optical depth for extinction of the layer, $\tilde{\tau}$:

$$d\tilde{\tau} = [1 - \omega_0(1 - b)] d\tau \quad (6.5)$$

which folds the scattering effects into the optical depth associated with atmospheric emission.

By using Eq. 6.4 and $\tilde{\tau}$, the equation for upwelling radiation observed from τ^* can be written as:

$$I^\uparrow(\tilde{\tau}^*, \mu) = I(\tilde{\tau}_{\text{surf}}, \mu) e^{(\tilde{\tau}^* - \tilde{\tau}_{\text{surf}})/\mu} + \int_{\tilde{\tau}^*}^{\tilde{\tau}_{\text{surf}}} B[T(\tilde{\tau})] e^{(\tilde{\tau}^* - \tilde{\tau}')/\mu} \frac{d\tilde{\tau}'}{\mu}. \quad (6.6)$$

which mimics the equation for upwelling radiance observed in clear-sky conditions using this new apparent optical depth.

Within RTTOVv12, $\tilde{\tau}$ is calculated using a rearrangement of Eq. 6.5:

$$d\tilde{\tau} = d\tau_a + b d\tau_s \quad (6.7)$$

where τ_a and τ_s are the optical depths associated with the absorbed radiation and backward-scattered radiation, respectively, and are calculated using the bulk absorption and scattering coefficients. The cloudy coefficients stored within RTTOVv12 contain these three spectrally dependent parameters (the bulk absorption coefficient, the bulk scattering coefficient, and the backscattering parameter), and they are derived from cloud optical property models.

In this work, the cloudy coefficients based on the Yang et al. (2013) and Baum et al. (2014) models are used given their wide usage in far-infrared studies (Bantges et al. 2020, Di Natale & Palchetti 2022). The Yang et al. (2013) and Baum et al. (2014) optical property models are defined for three different habits: the General Habit Mix (GHM), Aggregate Solid Columns (ASC), and Solid Columns (SC). While the SC and ASC models only include solid columns and aggregate solid columns, respectively, the GHM model incorporates plates, droxtals, hollow and solid columns, hollow and solid bullet rosettes, an aggregate of solid columns, and a small and large aggregate of plates. Only the GHM model has been converted into RTTOVv12 cloudy coefficients, and it is parameterised in terms of IWC from 0.5E-5 to 0.1 gm^{-3} and CER from 5-60 μm in each channel.

The use of the Chou approximation to simulate the cloudy radiances significantly increases the operational speed of the retrieval, as RTTOVv12 only needs to compute the clear-sky form of the radiative transfer equation with this apparent optical depth. However, the Chou approximation has been shown to introduce a positive bias into the spectrum within the far-infrared that is dependent on the cloud parameters. Martinazzo et al. (2021) showed that for an ice cloud composed of aggregate solid columns based on Yang et al. (2013), a COT of 1, a CER between 30-40 μm , and a CTH of 8 km in a mid-latitude atmosphere, this positive bias peaks at $\sim 1\text{K}$ at $\sim 410 \text{ cm}^{-1}$, which exceeds the FORUM-aircraft uncertainty by $\sim 0.4 \text{ K}$. A lower COT and larger CER reduce this bias. Within the mid-infrared, there is a negative bias that is approximately half the FORUM-aircraft uncertainty.

This smaller bias observed in the mid-infrared is because the spectral signatures of ice clouds within the atmospheric window are mostly dependent on the cloud's absorption features, rather than the scattering ones, which can be approximated more easily using the Chou approximation (Maestri & Holz 2009). In the far-infrared, a minimum in the complex component of the refractive index of ice results in larger scattering contributions in this spectral region. Simple scaling approximations, such as the Chou approximation, struggle to reproduce the heterogeneous angular variation of an incident radiance field associated with multiple scattering, causing these larger residuals (Martinazzo et al. 2021).

This motivates the first area of study and the first set of retrievals within this chapter, and the impact of the Chou approximation on the retrieval will be explored in Section 6.5.

6.2.2 Retrieval Approximations

During the retrieval process, RTTOVv12 ingests and outputs the IWC and CER on its 101 fixed levels, as well as their corresponding jacobians (stored in $\mathbf{K}_x$). The IWC and CER are translated into singular values of CER, COT, and CTH that are stored in the state vector for each iteration of the retrieval. The CER can be easily translated from RTTOVv12 into the state vector as the CER is assumed to be constant throughout the cloud. On the other hand, the COT and CTH are not explicitly output by RTTOVv12, and so are inferred from the vertical profile of the IWC.

At each iteration of the retrieval, the vertical profile of the IWC is assumed to be a Gaussian curve that is centred on a given altitude with a standard deviation of 1 km. The CTH is derived from 2 standard deviations above the centre of this Gaussian. Within IMS, the COT is derived from the IWC profile using the general approximation from Wen & Rose (1994) for all types of cloud:

$$\text{COT}_{\text{vis}} = \int_0^z \frac{3\text{IWC}(z) \langle Q_e \rangle}{4\rho\text{CER}} dz \quad (6.8)$$

where $\langle Q_e \rangle$ is the bulk extinction efficiency and the limits of the integral represent the geometrical thickness of the cloud. As the visible COT is being retrieved, it is assumed the average particle is much larger than the wavelength of incident radiation and so $\langle Q_e \rangle = 2$.

This approximation has been previously used to retrieve the COT of ice clouds, and so this approximation is also used in this work, but with $\rho = \rho_{\text{ice}}$ (Wang et al. 2011).

The jacobian for the COT ($\mathbf{K}_{\text{COT}}$) and the jacobian for the CTH ($\mathbf{K}_{\text{CTH}}$) must also be calculated so that perturbations can be applied to the state vector with each iteration (Eq. 2.30). These are calculated using the below relations:

$$\mathbf{K}_{\text{COT}} = \mathbf{K}_{\text{IWC}} \frac{d(\text{IWC})}{d(\text{COT})} \quad (6.9)$$

where the derivative of IWC with respect to COT is calculated using Eq. 6.8, and

$$\mathbf{K}_{\text{CTH}} = \mathbf{K}_{\text{IWC}} \frac{d(\text{IWC})}{d(\text{CTH})} \quad (6.10)$$

where the derivative of IWC with respect to CTH is derived from the vertical profile of the IWC.

6.3 B895 Flight Overview

The B895 flight took place on 13 March 2015 as part of the CIRrus Coupled Cloud-Radiation EXperiment (CIRCCREX) campaign which had the overarching goal of improving our understanding of cirrus cloud microphysics. The main objective of the flight was to simultaneously measure upwelling radiances in both the mid- and far-infrared using TAFTS (Canas et al. 1997), and ARIES (Wilson et al. 1999) above a well-characterised cirrus cloud. These radiance observations were collected by Jonathan Murray (Imperial College London), Cathryn Fox (Imperial College London), and Stuart Fox (Met Office). The flight was performed using the FAAM aircraft and was based out of Prestwick, Scotland.

Figure 6.2 shows the flight track overlaid on top of the Level 2 COT from the MODIS Terra overpass at 10:40 UTC, with notable timings highlighted in Table 6.1 for clarity. The FAAM aircraft performed three SLRs at an altitude of 9.4 km, overflying a decaying band of cirrus cloud associated with an occluded front. This work focuses on a radiance observation taken at 09:48:39 UTC during the first SLR (09:33 to 09:58 UTC), where the plane was flying in a north-westerly direction. At this time, the atmosphere above the aircraft was cloud-free,

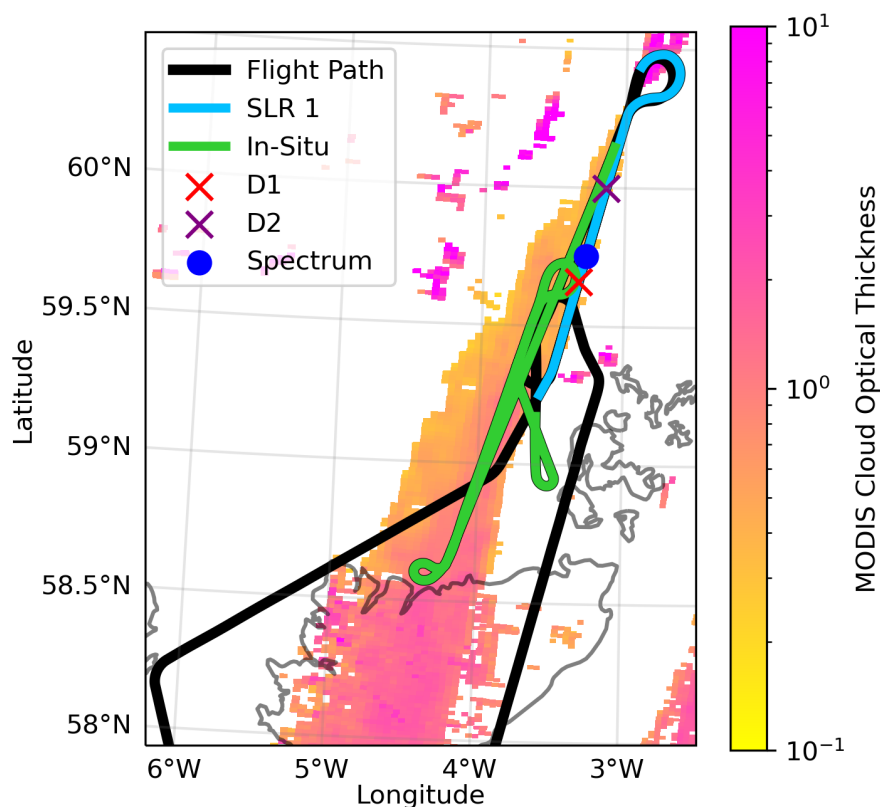


Figure 6.2: The flight track (black) for the B895 flight that took place on 13 March 2015 plotted on the cloud optical thickness product from the MODIS Terra satellite overpass at 10:40 UTC. The first straight and level run (SLR 1) is shown in light blue, and the location of the selected radiance observation is marked with a blue circle (09:48:39 UTC). Dropsondes released during SLR 1 are marked with a cross. Approximately 50 minutes after the radiance observations were taken, the aircraft descended through the cloud, taking in-situ measurements following the light green path between 10:32 and 11:32 UTC.

Table 6.1: The timings of relevant measurements taken during the B895 flight.

	Time (UTC)
SLR 1	09:33 – 09:58
Radiance Observation	09:48:39
Dropsonde 1 Release	09:47:48
Dropsonde 2 Release	09:50:42
In-situ Measurements	10:32 – 11:32
MODIS Terra Overpass	10:40

limiting the amount of downwelling radiation available for backscatter into the radiometric instrumentation from the underlying cirrus. Following the method outlined in Chapter 3, this radiance observation (1 TAFTS and 6 ARIES scans) has been converted into a FORUM-aircraft observation (Figure 6.3) to mimic the FSI spectral characteristics. As a result, it

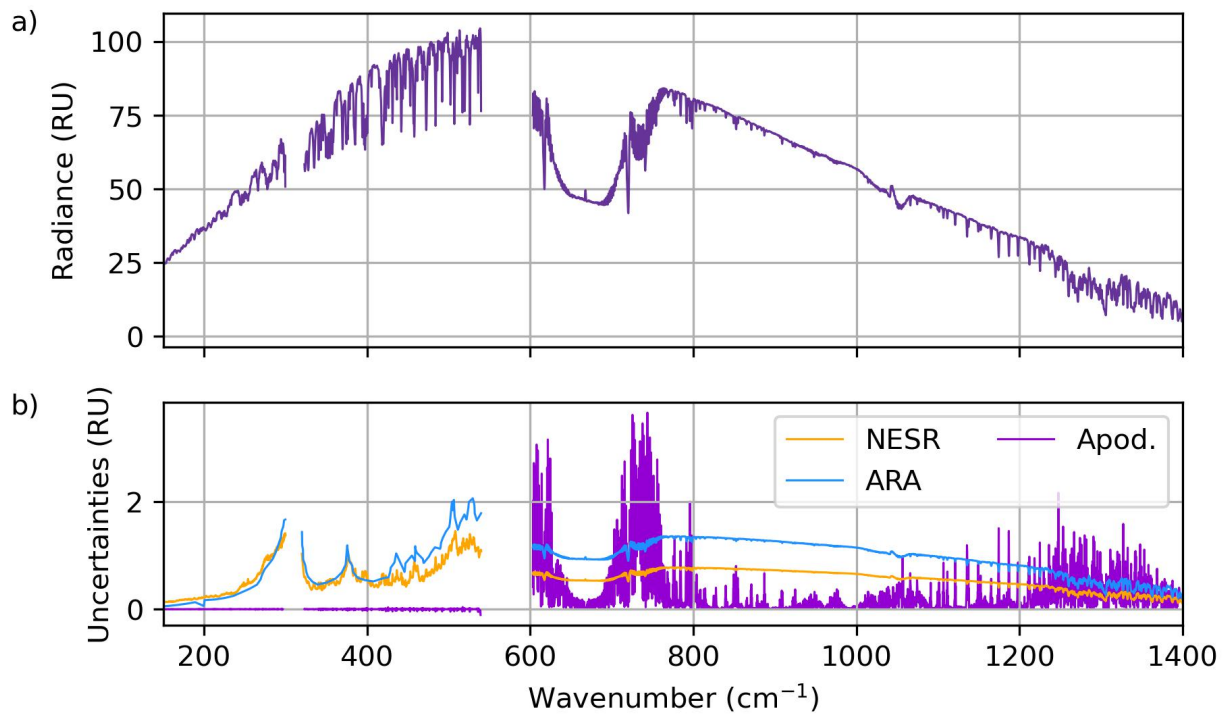


Figure 6.3: (a) The FORUM-aircraft observation taken at 09:48:39 UTC during the first SLR. The spectral gaps correspond to the separation between the TAFTS shortwave and longwave channel, and the separation between the TAFTS shortwave and ARIES longwave channel. (b) The uncertainties associated with the FORUM-aircraft observation. The NESR (orange) corresponds to one TAFTS scan and six ARIES scans, and is based on the single scan NESR shown in Figure 3.3b. The ARA (blue) for TAFTS and ARIES are the same as in Figure 3.3c. The apodisation (Apod.) uncertainty (violet) is explored in Chapter 3 and is the same as the violet line in Figure 3.6. The radiance units are $\text{mW}/\text{m}^{-2}/\text{sr}^{-1}/\text{cm}^{-1}$ and are used for both subplots.

has an increased uncertainty in the $15 \mu\text{m}$ CO_2 wings and above 1200 cm^{-1} . This is shown in radiance units in Figure 6.3b alongside the NESR and ARA of the instruments. It was originally presented in Figure 3.6. The NESR, ARA, and apodisation uncertainty are combined in quadrature to estimate the uncertainty in the FORUM-aircraft observations (the FORUM-aircraft uncertainty).

The aircraft was also equipped with the Airborne Vertical Atmospheric Profiling System, which released two Vaisala RD94 dropsondes at 09:47:48 and 09:50:42 UTC during the first SLR. These are the same type of dropsondes described in Section 5.2.1, and so it is estimated that they have the same uncertainties for temperature and water vapour. To summarise, the uncertainty in the temperature profile measured by the dropsonde was 0.2 K. There was a calibration uncertainty of $\pm 5 \%$ in relative humidity with an offset of $\pm 0.5 \%$, and a

production uncertainty of ± 1.5 % in relative humidity above 10 % and ± 3 % below 10 % relative humidity.

After completing the other two SLRs, the plane descended through the cloud and measured the microphysics using on-board instrumentation between 10:32 and 11:32 UTC. During these manoeuvres, the cirrus remained relatively stationary, with satellite imagery indicating that it had dispersed by 13:00 UTC (O'Shea et al. 2016).

6.3.1 Cloud Instrumentation

There were a number of instruments onboard the FAAM aircraft used to characterise the cirrus cloud that will be used to assess the retrievals performed in this thesis.

A Leosphere ALS450 355 nm elastic backscatter lidar was installed on the aircraft (Marenco 2010). This provided information on the cloud vertical extent, ice volume extinction profiles at 355 nm from the range-corrected backscatter profiles following the two-stage process of Marenco et al. (2011), and a vertical profile of the particle extinction coefficient using the method described in Fox et al. (2019). Raw lidar profiles were recorded with a vertical resolution of 1.5 m and an integration time of 2 s. The range-corrected backscatter profile measured by the lidar at 09:40:35 UTC (4 seconds before the radiance observation) is shown in Figure 6.4a alongside the lidar-derived extinction profile (Figure 6.4b), and the temperature and water vapour profiles measured by dropsonde 1 (Figure 6.4c and d). This lidar-derived extinction coefficient profile has been previously used to derive the COT, however, due to poor instrument alignment, there were issues with the depolarisation calibration which limited the quality of the estimated COT (Fox et al. 2019).

To mitigate these issues, a simplified retrieval approach has been developed by Rui Song (University of Oxford) to derive the COT rather than the full vertical extinction profile from the lidar measurements. The finalised COT from the B895 flight was calculated by Rui Song and provided to support this work. This approach considers the power of the attenuated Rayleigh backscatter range-corrected signals below 6 km in cirrus-free (P_{ref}) and cirrus (P_{att}) conditions. It is assumed that the molecular extinction is constant along the short flight path, and so the signal variation can be attributed to the ice crystals in the cirrus cloud. The

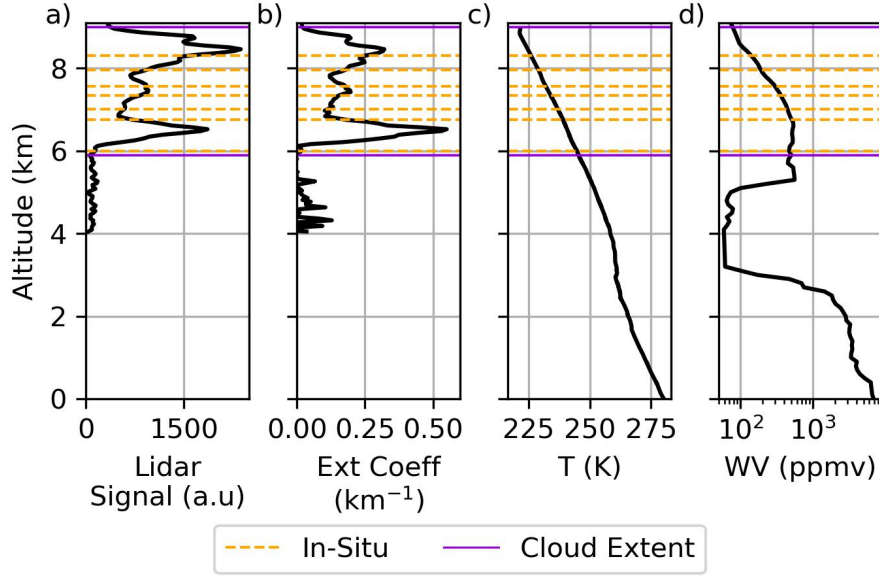


Figure 6.4: (a) The range-corrected lidar signal of the cloud measured at 09:48:35 UTC. (b) The lidar-derived extinction coefficient of the cloud measured at 09:48:35 UTC. The (c) temperature and (d) water vapour profiles measured by dropsonde 1 at 09:47:48 UTC. The uncertainty in the dropsonde measurements are described in Section 6.3 and are estimated to be 0.2 K and $\sim 10\%$ for temperature and water vapour, respectively. The edges of the cloud extent are marked in purple, and the altitudes at which in-situ measurements of cloud habit and PSDs have been characterised are shown with dashed orange lines (6, 6.8, 7, 7.3, 7.6, 8.0, and 8.3 km).

presence of cirrus reduces P_{att} relative to P_{ref} through two-way transmission and simplifies the COT retrieval to:

$$\text{COT}_{355 \text{ nm}} = -\frac{1}{2} \ln \left(\frac{P_{\text{att}}}{P_{\text{ref}}} \right) \quad (6.11)$$

where the factor of 2 accounts for the two-way transmission through the cirrus layer. In this work, P_{ref} is calculated from the average of one minute of backscatter signals between 4 and 6 km at 09:33 UTC where the atmosphere was cloud-free below the aircraft. P_{att} represents the backscatter signals from the remainder of the SLR that have then also been vertically averaged between 4 and 6 km for each of the 2 s measurement intervals. This means the average lidar-derived COT for the one-minute period surrounding the radiance observation is stated as 0.8 ± 0.1 , with the uncertainty representing the standard error in the mean COT within this time period.

The PSDs and ice crystal habits were measured using a series of probes that included

a Cloud 2-DS probe, a 3-View Cloud Particle Imager (3V-CPI), a Cloud Imaging Probe 100 (CIP 100) and a holographic cloud probe (HALOHolo) (Lawson et al. 2006, O'Shea et al. 2016). The Cloud 2-DS and inlet edge of the 3V-CPI probes were fitted with antishatter tips to reduce a bias towards smaller crystals. O'Shea et al. (2016) presented an analysis of the in-situ measurements taken during the B895 flight, with both PSDs and habits divided into the different temperature regions shown in Figure 6.4. Each set represents the average of runs through sections of the cloud, each taking between 5 and 10 minutes. The best-fit distributions for the PSDs were generally found to be bimodal in O'Shea et al. (2016), and these will be used to estimate the CER in Section 6.3.2.

The habits of particles larger than $50 \mu\text{m}$ were determined from the 3V-CPI images using an automatic habit recognition algorithm outlined in O'Shea et al. (2016). Particles were found to be a mix of predominantly aggregates and droxtals, with some rosettes and columns. This is more similar to the constituents of the GHM model than the SC model, and so it is expected that the Yang et al. (2013) and Baum et al. (2014) GHM model will be a better representation of the ice crystal habits within this cirrus cloud.

6.3.2 Estimating the CER

The in-situ measurements of the cloud PSDs and ice crystal habits analysed in O'Shea et al. (2016) can be used to provide an approximation of the CER. The CER used in the Yang et al. (2013) and Baum et al. (2014) bulk optical property models is defined using Eq. 2.13 and is a ratio of the total volume and projected area of the ice crystals within the bulk cloud. These are calculated using Eq. 2.15 and 2.14, respectively.

Figure 6.5 shows the particle size distributions derived from the Cloud 2-DS probe that were published in O'Shea et al. (2016) in black. Following the findings of O'Shea et al. (2016), the number distribution at each altitude is found by fitting the PSDs measured by the Cloud 2-DS probe with bimodal Gaussian distributions using the non-linear least squares method (Zhao et al. 2011, Jackson et al. 2015). The successfully fitted distributions are shown as dashed red lines in Figure 6.5. Due to the dominance of smaller crystals, as observed in O'Shea et al. (2016), the PSDs measured at 232 and 226 K (7.5 and 8.3 km) could not be fit

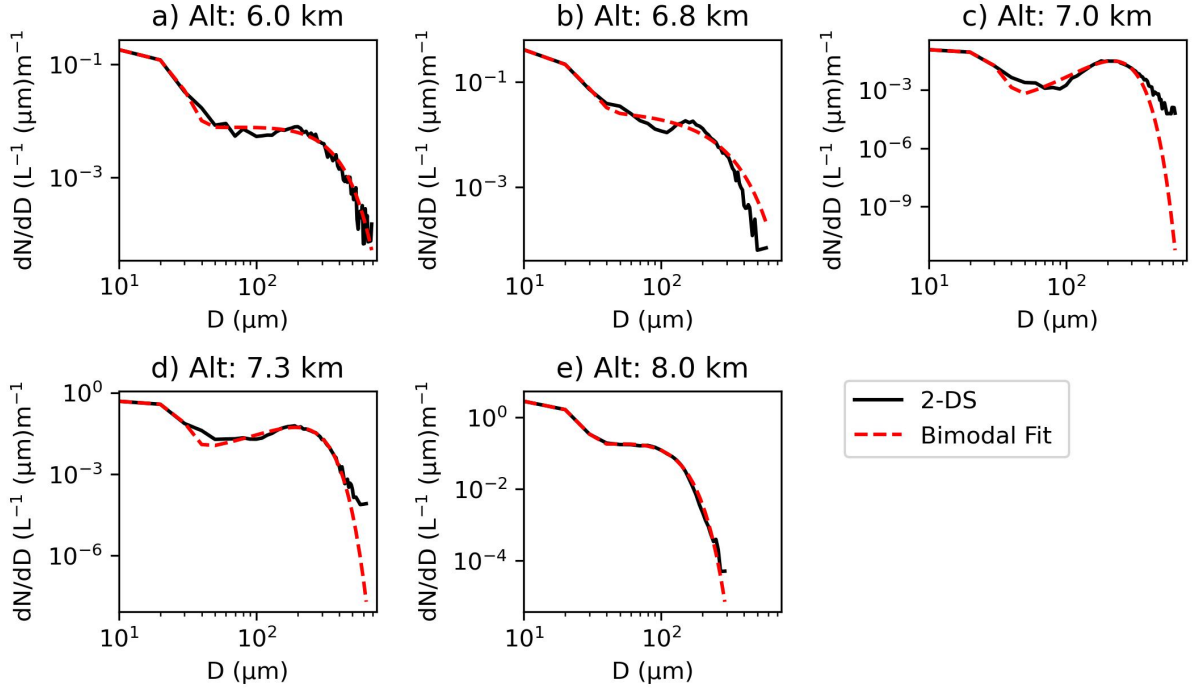


Figure 6.5: The (black) particle size distributions measured by a Cloud 2-DS probe during the B895 flight on 13 March 2015 that were successfully fitted with a bimodal Gaussian distribution.

Table 6.2: The relative standard deviation (%) in each parameter fitted in the bimodal Gaussian: $f(x) = A_1 \exp \left[\frac{-(x-\mu_1)^2}{2\sigma_1^2} \right] + A_2 \exp \left[\frac{-(x-\mu_2)^2}{2\sigma_2^2} \right]$ corresponding to the red dashed curves in Figure 6.5.

Altitude (km)	A_1	μ_1	σ_1	A_2	μ_2	σ_2
6.0	3	3	0.8	50	10	6
6.8	40	7	5	100	40	100
7.0	2	4	2	0.7	2	2
7.3	2	3	1	2	4	3
8.0	0.8	1	0.3	4	3	1

with a bimodal Gaussian distribution, and so they have been excluded from this work.

The habits and habit fractions were derived from the 3V-CPI measurements and the results of the habit recognition algorithm presented in O'Shea et al. (2016). The area and volume of each crystal with a given habit and size were derived from the Yang et al. (2013) database. The uncertainty presented here for the CER is calculated by combining the relative standard deviation of the parameters (Table 6.2) used to fit the PSDs, and so represents one standard deviation in the fitting of each bimodal Gaussian distribution to each PSD.

Table 6.3: The CER calculated from the PSDs measured by the Cloud 2-DS probe, and the habit distributions measured by the 3V-CPI for temperature regions shown in Figure 6.4. The temperatures have been converted to altitudes using the Dropsonde 1 measurements (Figure 6.4c) and have been used to calculate the approximate times and distances since and from the radiance observation.

Altitude (km)	Time Since Rad Obs (min)	Dist to Rad Obs (km)	CER (μm)
6.0	95	71	42 ± 5
6.8	80	38	47 ± 6
7.0	75	98	29 ± 1
7.3	65	126	20 ± 1
8.0	55	72	56 ± 1

It is important to note that there is potentially a systematic bias in the smaller mode of the PSDs that would increase the uncertainty in the CER. O'Shea et al. (2021) found that in an earlier CIRCCREX flight (11 March 2015) the smaller mode of each PSD measured by the Cloud 2-DS probe was increased by the ice crystals diffracting the incoming beam and creating shadows on the photodiodes of both probes. This diffraction effect was reduced by filtering the interarrival times between the two stereo probes in O'Shea et al. (2021), however, it did not fully eliminate the bias in all cases. A similar quality control was performed in O'Shea et al. (2016) on the PSDs, and so it is likely that the uncertainties presented in Table 6.3 are an underestimation of the true uncertainties in the derived CER from each PSD.

Table 6.3 shows the estimated CER values for each altitude with approximate times and distances relative to that of the radiance observation. These range from 20-56 μm with an uncertainty of up to 12 % that increases closer to the base of the cloud, and were taken up to 95 minutes after the time of the radiance observation. The mean CER weighted by the individual uncertainties is 32 μm with a weighted standard deviation of 14 μm that represents the spread of CER measured within the cloud.

6.4 Optimising IMS for Ice Cloud Retrievals from the B895 Flight

This section details the changes that have been made to IMS to optimise retrievals of COT, CER, CTH, ice crystal habit, temperature, and water vapour from the FORUM-aircraft radiances taken during the B895 flight.

6.4.1 Constructing Other Ice Crystal Habit Coefficients

As described in Section 6.2.1, the Yang et al. (2013) and Baum et al. (2014) cloud optical models are provided for three different habit models: GHM, ASC, and SC. Currently, only the GHM model has been parameterised into the RTTOVv12 cloudy coefficients and is available on the RTTOVv12 website. Given the additional information stored within the far-infrared about ice crystal habit, this section builds cloudy coefficients from the ASC and SC models.

Each of the optical property models includes the bulk extinction coefficient, bulk scattering albedo, and phase functions (for 0 to 180 deg) defined in wavelengths from 0.2 to 99 μm and for CER from 5-60 μm . Using Eq. 2.17 from Chapter 1, the bulk absorption and scattering coefficients can be calculated, and equations 6.3 and 2.12 can be used to calculate the backscattering parameter (Saunders et al. 2017). Exact details of how these parameters are translated into wavenumber space and onto the FSI spectral grid were not provided by the RTTOVv12 team with the existing GHM coefficients. Therefore, as an approximation, these parameters have been interpolated to the spectral sampling of the FSI instrument in wavenumber space. Only three FORUM-aircraft channels are outside the spectral range of this optical property model (100, 100.3, and 100.6 cm^{-1}), and so will be excluded from the retrieval.

This method of parametrization is first tested on the GHM model and compared against the original GHM cloudy coefficients provided with RTTOVv12. The percentage differences between the new and original RTTOVv12 coefficients are shown in Figure 6.6. As the models are originally defined in wavelength, the interpolation to the FSI wavenumber scale leads to larger percentage differences within the far-infrared, but these remain below 3 %. Different

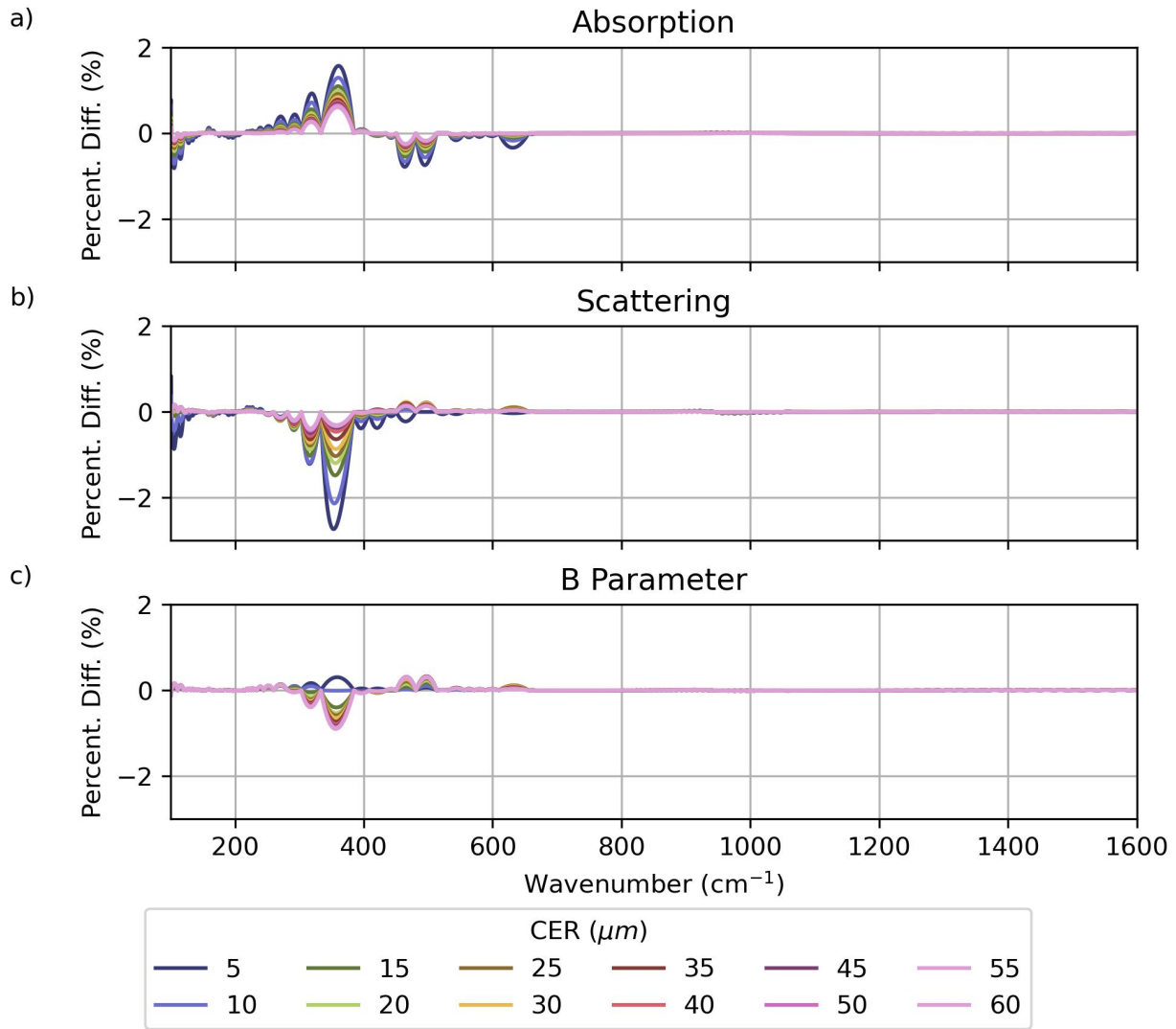


Figure 6.6: The percentage difference between the original RTTOVv12 cloudy coefficients, and the ones built in this work for the (a) absorption and (b) scattering coefficients, as well as the (c) backscattering parameter (B Parameter). Differences are shown for the CER limits within the cloud optical property models ranging from (5-60 μm).

methods of interpolation were tested to minimise this difference, but it was found that the coefficients derived using the linear interpolation (shown here) had the smallest percentage differences to the original coefficients.

The impact of the percentage differences shown in Figure 6.6 on the simulated radiances was tested by performing RTTOVv12 simulations that used the vertical temperature and water vapour in Figure 6.4c and d, an IWP of 28 gm^{-2} , and a CER that varied from 5-60 μm in increments of 5 μm . The residuals (not shown) between RTTOVv12 simulations that used the original and newly constructed GHM coefficients were of the order of $0.001 \text{ mW/m}^2/\text{sr/cm}^{-1}$

which is significantly smaller than the FORUM-aircraft uncertainty (components in Figure 6.3b). Therefore, it is assumed that the ASC and SC bulk optical property models can now also be converted into cloudy coefficients and be used to simulate cloudy radiances.

Implications on the Retrieval Capability

In order to retrieve the ice crystal habit with any confidence, the radiance residuals between simulations using the different habits must exceed the FORUM-aircraft uncertainty. Bantges et al. (2020) highlighted the radiance residuals between LBLDIS (LBLRTMv12.8) spectra simulated using each of the three optical property models. LBLDIS was introduced in this thesis in Section 2.1.3. It uses numerical methods to calculate the contributions of ice crystal scattering to the upwelling radiances, and is considered one of the most accurate models for simulating multiple scattering. In each LBLDIS simulation in this section, the same temperature, gaseous and surface, CER, and bulk extinction coefficient profiles were used to simulate the radiance observation taken at 09:49:39 UTC.

The temperature and water vapour profiles were derived from dropsonde 1, and the O_3 profile from co-located ERA5 data. Profiles for CO_2 , CH_4 , and other trace gases were set to the US 1976 mid-latitude winter profile scaled to present-day concentrations using data from Mace Head (Dlugokencky et al. 2019). The surface skin temperature was set to 280.2 K, which was derived from the dropsonde profile and co-located ERA-I data. The spectral surface emissivity was calculated using the Masuda et al. (1988) model above 769 cm^{-1} . Below 769 cm^{-1} , the surface emissivity was found to have a negligible effect on the spectrum, given the strong water vapour absorption below the cloud in the far-infrared (Bantges et al. 2020). Therefore, it was fixed to 0.99. Based on the results of the minimisation process in Bantges et al. (2020), the lidar-derived extinction coefficient profile (Figure 6.4a) was scaled to output a COT_{355nm} of 0.82, and the CER was set to $34\text{ }\mu\text{m}$. The residual between this FORUM-aircraft simulation and the FORUM-aircraft observation is shown in Figure 6.7. It is important to note that in this Chapter the residuals between the retrieved and observed spectra are presented in brightness temperature, as this can be useful for interpreting the implications on the retrieved CTH in the context of the temperature profile. As a result, the residuals between the

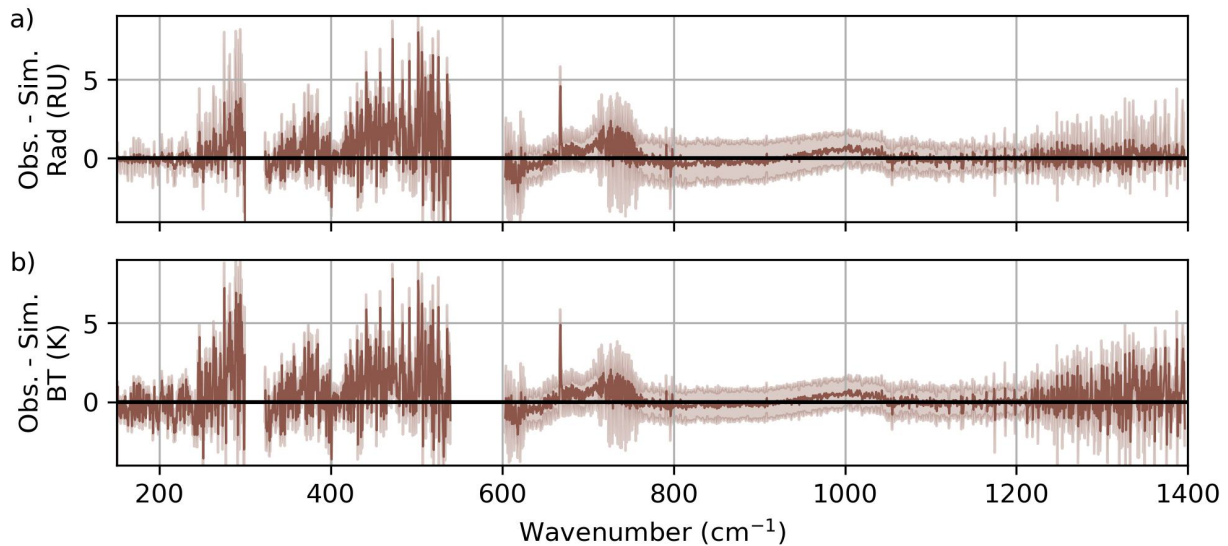


Figure 6.7: The residual between the FORUM-aircraft observation (Figure 6.3a) and the FORUM-aircraft simulation described in Section 6.4.1 given in (a) radiance (Rad) with radiance units (RU) of $\text{mW}/\text{m}^2/\text{sr}/\text{cm}^{-1}$ and (b) brightness temperature (BT). The shading represents the uncertainty in the FORUM-aircraft observation and is composed of the instrument noise, calibration uncertainty, and uncertainty introduced through the FORUM-aircraft apodisation added in quadrature. Both units are given to ease comparisons made throughout this Chapter.

simulated and observed FORUM-aircraft spectra are presented in radiance units (Figure 6.7a) and brightness temperature (Figure 6.7b) to aid later analysis. The simulation is generally within 5 K (or $5 \text{ mW}/\text{m}^2/\text{sr}/\text{cm}^{-1}$) of the observation, excluding selected channels near the TAFTS band edges, and will be used to explore the expected behaviour of the retrieval.

The radiance residuals from Bantges et al. (2020) have been reproduced in Figure 6.8a using LBLDIS (LBLRTMv12.11) with the FORUM-aircraft spectral characteristics, and are plotted against the observation's uncertainty. The radiance residuals between LBLDIS spectra simulated using the GHM and SC or ASC habit do not exceed the FORUM-aircraft uncertainty in the mid-infrared, suggesting the difference between these habits would not be detectable when using the mid-infrared alone. The FSI target uncertainty is also plotted for comparison. While the difference between the GHM and ASC simulations still does not exceed the FSI target uncertainty in the mid-infrared, the difference between the GHM and SC simulations does between $\sim 700\text{-}900 \text{ cm}^{-1}$ and at $\sim 1000 \text{ cm}^{-1}$. However, both sets of radiance residuals exceed the FORUM-aircraft (and target FSI) uncertainty in the far-infrared, suggesting that this region could be exploited to improve our understanding of ice crystal habits.

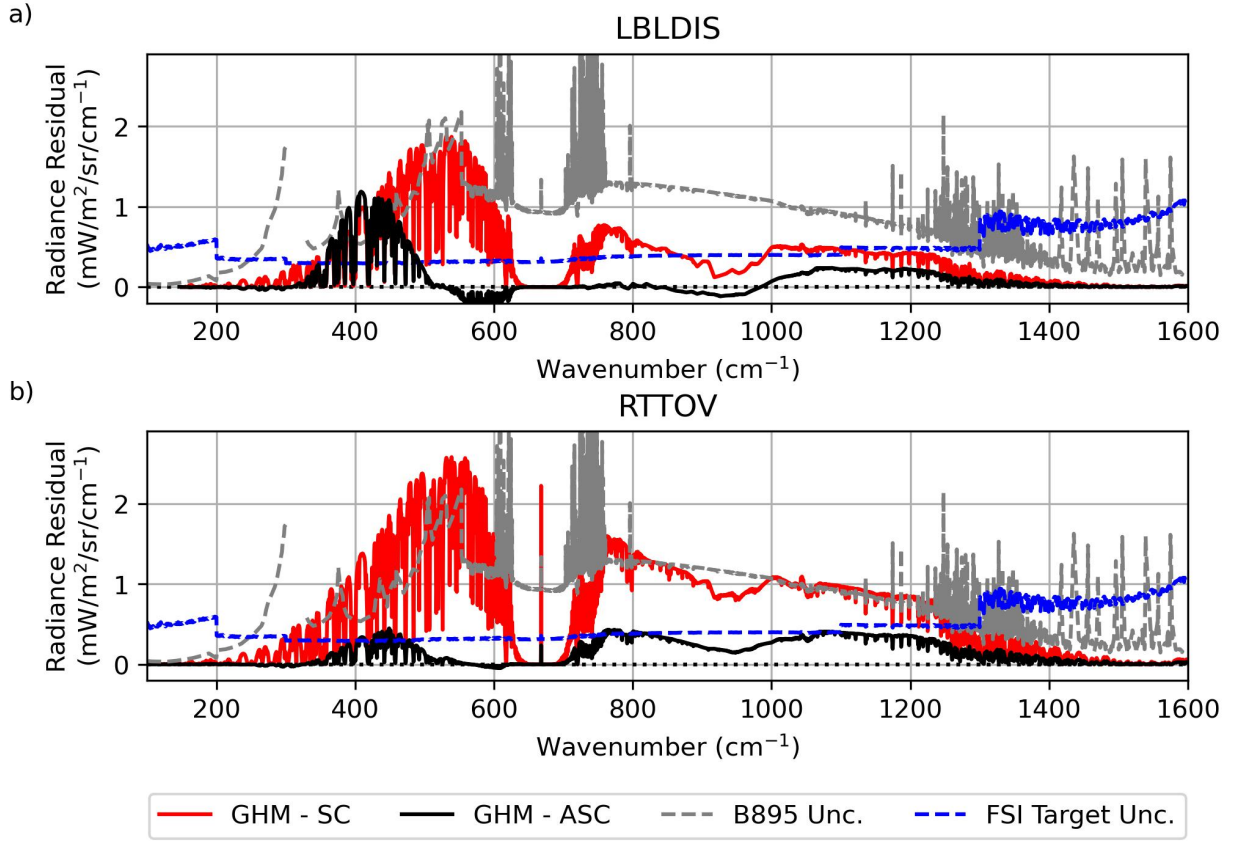


Figure 6.8: The radiance residuals between the GHM and SC model (red) and the GHM and ASC model (black) are shown for the (a) LBLDIS and (b) RTTOVv12 models. The vertical profile of temperature and water vapour, and ice crystal properties are taken from Bantges et al. (2020), and are based on the profile in Figure 6.4 with a CER of $34 \mu\text{m}$ and a CTH of 9 km used in both models. A COT of 0.82 is used in LBLDIS and an IWP of 28 gm^{-2} is used in RTTOVv12. The FSI target uncertainty for this case is shown in blue. The uncertainty associated with the FORUM-aircraft observation is shown in grey, and is composed of the instrument noise, calibration uncertainty, and the uncertainty associated with the FORUM apodisation process described in Chapter 3 added in quadrature. Each of these components were presented in Figure 6.3b.

A similar evaluation of the radiance residuals using the different habits has been performed using RTTOVv12. While these simulations use the same temperature, gaseous, and CER profile as in Figure 6.8a, RTTOVv12 defines the cloud using CER and IWC rather than the CER and bulk extinction coefficient inputs used in LBLDIS. Therefore, the vertical profile for the IWC has been taken from Bantges et al. (2020), and was derived from the lidar measurements following adjustments made to the backscatter-to-extinction ratio. It has the same vertical shape as the extinction coefficient in Figure 6.4a and totals an IWP of 28 gm^{-2} .

A direct comparison between LBLDIS and RTTOVv12 is not performed in this thesis

given the difference in inputs for either model, and an evaluation of the impact of the Chou approximation (assuming the ASC habit) has already been performed in Martinazzo et al. (2021). Therefore, the RTTOVv12 radiances residuals caused by different habits are only considered here as a first step in determining if IMS can be used to perform retrievals of ice crystal habit from this specific FORUM-aircraft observation.

The RTTOVv12 radiance residuals (Figure 6.8b) between the GHM and SC models exceed the FORUM-aircraft uncertainty at $\sim 790 \text{ cm}^{-1}$, and $\sim 1200 \text{ cm}^{-1}$ in the mid-infrared, but are up to $\sim 1 \text{ mWm}^{-2}\text{sr}^{-1}/\text{cm}^{-1}$ larger in the far-infrared. This indicates a greater sensitivity to these ice crystal habits that can be used to distinguish between the GHM and SC habits when including far-infrared channels in the retrieval. However, the maximum radiance residuals between the GHM and ASC models are $\sim 1 \text{ mWm}^{-2}\text{sr}^{-1}/\text{cm}^{-1}$ smaller when simulating using RTTOVv12 in comparison to LBLDIS and are only slightly larger than the FORUM-aircraft uncertainty. This suggests that when using RTTOVv12, the difference between GHM and ASC habits could be less detectable in retrievals from the FORUM-aircraft observation analysed in this chapter. Therefore, for the remainder of this chapter, only the GHM and SC habits are considered.

6.4.2 Retrieving the Ice Crystal Habit

Unlike the other retrieval targets, the ice crystal habit will be retrieved by examining the spectral fit in certain channels after the optimal estimation retrieval of the other targets has been performed. To do this, the BT residuals in individual channels and the measurement cost per channel (J_y) are considered, enabling comparisons between different configurations. A lower J_y for a given habit or habit mix is indicative that it is a better representation of the true crystal shapes within the cloud. If $J_y < 1$, then the retrieval has fit the spectra on average within the uncertainty. For guidance, an increase of 20 % in J_y is indicative of a 10 % increase in the residual between the observed and fitted spectra, assuming the uncertainty is kept constant.

In each set of retrievals, there are 4 different configurations used: with and without the far-infrared and assuming the SC or GHM bulk optical property model. These will be referred

Table 6.4: The 4 different retrieval configurations used: with and without the far-infrared, and assuming the SC or GHM bulk optical property model.

Name	MIR Channels	Far-infrared Channels	Habit
MIR (GHM)	X		GHM
MIR (SC)	X		SC
MIR+FIR (GHM)	X	X	GHM
MIR+FIR (SC)	X	X	SC

to using the names outlined in Table 6.4 for clarity.

As with Chapter 5, the ESD and DOFS are used to evaluate the retrieval. Similarly, the temperature and water vapour profiles measured by the dropsonde are treated with the corresponding averaging kernels (Eq. 2.43), and are referred to as AK-treated dropsonde profiles.

6.4.3 Optimal Estimation Retrieval Constraints

In this work, optimal estimation retrievals of temperature, water vapour, COT, CER, and CTH are performed using both the GHM and SC ice crystal habit models. The default a-priori and covariance for temperature and water vapour are used (derived from global ERA5 across three days). COT is stored in log form within the state vector to prevent it from reducing below zero, and it is assumed there is minimal cloud at first, resulting in an a-priori of $\ln(0.01) \pm 10$. The default a-priori and covariance for the CTH is 7 ± 5 km, and for the CER is 30 ± 10 μm to represent the variability in ice, water, and mixed phase clouds. As this retrieval is only focused on cirrus clouds, the a-priori and covariance have been changed for CTH to 9 ± 3 km, to capture the higher altitude of cirrus clouds and for CER to 40 ± 10 μm due to the larger crystal sizes based on the climatology in Hong et al. (2016). These constraints were found to maximize the DOFS for the cloud retrieval targets for this case study.

The measurement covariance (Figure 6.9) is assumed to be fully correlated and is composed of the noise and calibration uncertainty for a single TAFTS and 6 ARIES scans with the apodisation uncertainty described in Chapter 3. Each of these components are presented in Figure 6.3b.

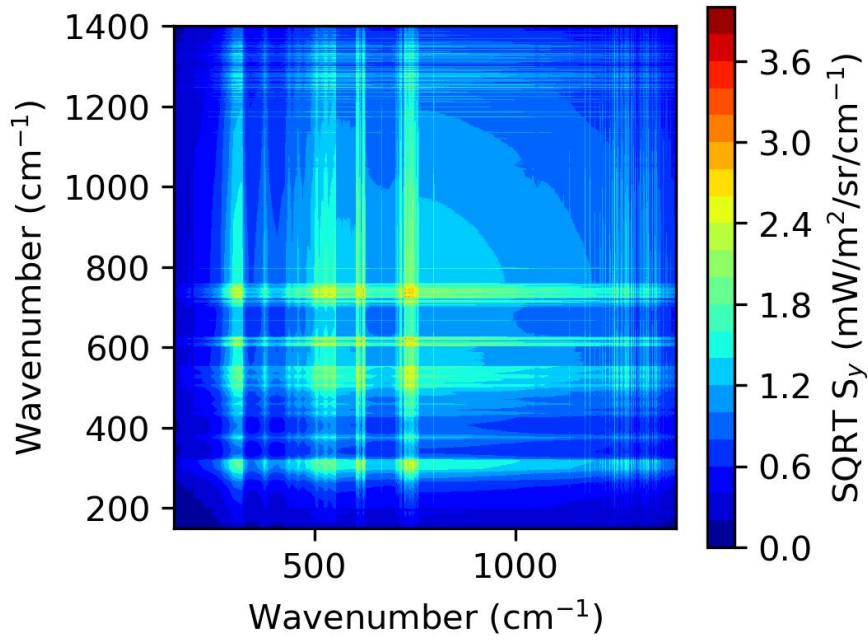


Figure 6.9: The square-root of S_y used in the retrievals in radiance units. This is a combination of the instrument NESR, ARA, and uncertainty introduced through the FORUM-aircraft apodisation process shown individually in Figure 6.3b.

6.4.4 Channel Selection

Figure 6.10 shows the 200 channels selected in Chapter 5 to optimise retrievals of temperature and water vapour from FORUM-aircraft observations. The absorption of water vapour dominates the far-infrared, and as a result, there is a significant overlap in this region between the spectral sensitivity to tropospheric water vapour and ice cloud properties. As a result, a number of the far-infrared channels that are sensitive to ice crystal habit have already been included for the water vapour retrieval. To enable a clearer distinction between habits, 26 additional channels have been included that are sensitive to ice cloud properties for this specific observation (Figure 6.10). It is anticipated that they are also sensitive to water vapour and temperature.

In Section 4.7, the channels used in the clear-sky retrievals were selected by exploiting the information content. The information content is a combination of the a-priori covariance that shows the variability of the retrieval targets, and the measurement covariance, which represents the uncertainty associated with simulating the measurement. This method is not applicable here because the a-priori covariance for the cloud retrieval targets was selected to

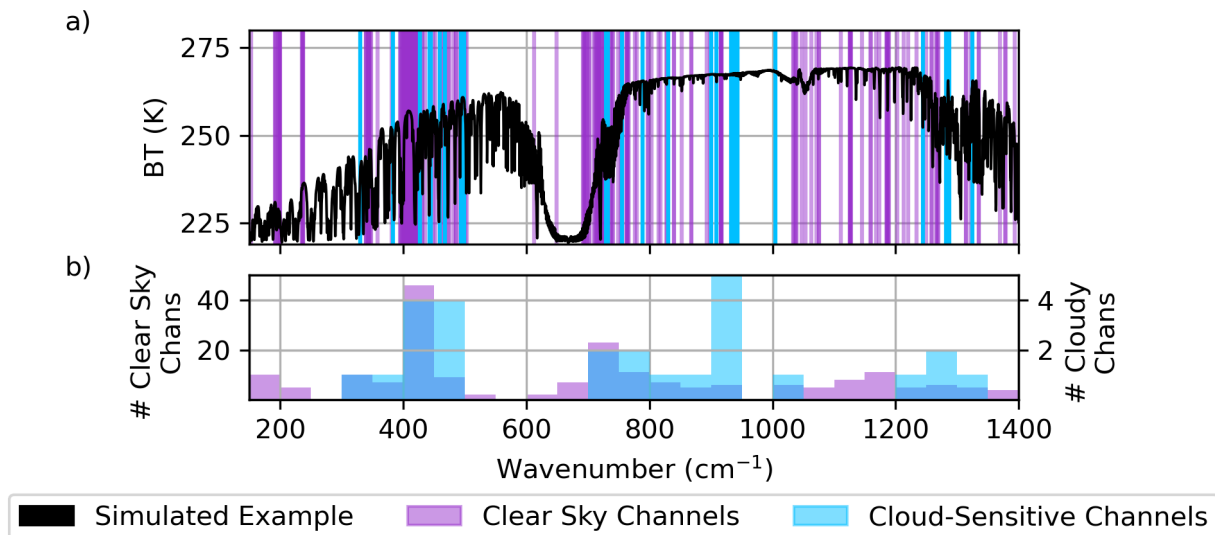


Figure 6.10: (a) The channels selected for retrievals using the FORUM-aircraft configuration of IMS on top of the FORUM-aircraft simulation in brightness temperature (BT), with (b) the number of channels in each 50 cm^{-1} bin. The 200 channels selected in Chapter 5 for optimised retrievals of temperature and water vapour are shown in purple. The additional 26 channels that have been selected to optimise retrievals of ice cloud properties are shown in blue.

be representative of ice, water, and mixed-phase clouds, rather than only ice clouds. Therefore, these additional ‘cloud-sensitive’ channels were selected using a database of FORUM-aircraft spectra simulated using RTTOVv12 that vary in COT (0.1-2), CER ($5\text{-}60 \mu\text{m}$), and ice crystal habit (GHM and SC models) but all have the same temperature, gaseous and surface profile as in the FORUM-aircraft simulations in Section 6.4.1. The channels not used in the clear-sky retrieval were first filtered based on whether clear-sky residuals between LBLRTMv12.11 and RTTOVv12 were comparable to, or exceeded the instrument uncertainty as was done in Chapter 5. This was to ensure that the uncertainty associated with the forward model could be directly attributed to its cloud assumptions. For each cloud property, these channels were then filtered based on two conditions. The first was that the largest changes in radiance were detectable above the FORUM-aircraft uncertainty. The second was that the radiance change was not perfectly correlated (i.e. a correlation coefficient of less than 1) to another channel. Channels with the largest change in radiance were selected first.

Figure 6.10 shows that 16 mid-infrared and 10 far-infrared additional channels were selected to improve the retrieval of ice cloud properties. To highlight the channels that are most sensitive to ice crystal habit, Figure 6.11 shows the BT residuals within these channels

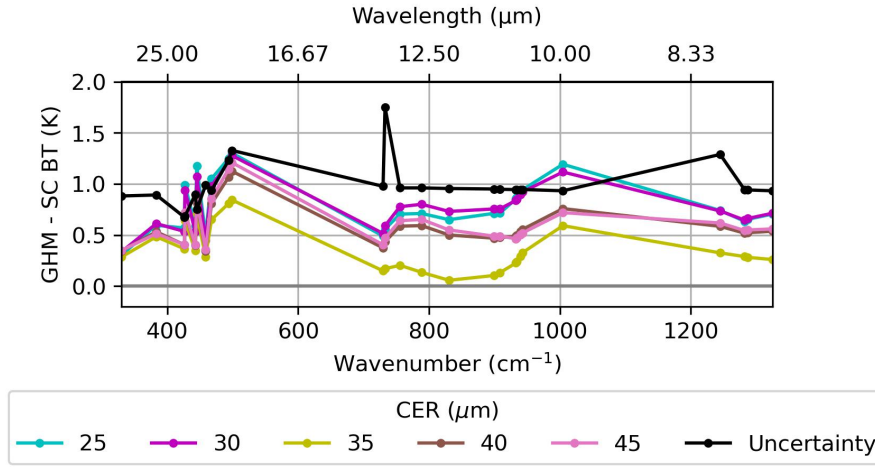


Figure 6.11: The brightness temperature (BT) residuals between the RTTOVv12 FSI simulated spectra assuming the GHM and SC databases for example CER values and an ice water path of $\sim 28 \text{ gm}^{-2}$ derived from the lidar measurements. The remainder of the atmospheric and surface profile are as described in Section 6.4.1. The uncertainty is square-root-diagonal of the measurement covariance in Figure 6.9, and is equivalent to the FORUM-aircraft uncertainty.

between the GHM and SC models for five different CER and a fixed IWP. Residuals between the models reach up to 1.3 K and exceed the FORUM-aircraft uncertainty between 400-450 cm^{-1} and in the 1004.5 cm^{-1} channel, and so it is to be expected that the habit will only be detectable within these channels for this case.

6.5 Fixed Temperature and Water Vapour Retrievals

Given the uncertainties and approximations associated with modelling the radiative interactions of ice clouds across the far-infrared, two initial retrievals of COT, CER, CTH, and ice crystal habit have been performed, assuming a fixed temperature, gaseous, and surface profile. The temperature and water vapour vertical profiles were fixed to the profiles measured by Dropsonde 1 (Figure 6.4c and d), while the remaining gaseous and surface profiles have been fixed to the values specified in Section 6.4.1. Only the cloud-sensitive channels are used in this test as this work is only focused on the cloud retrieval targets at this time. Therefore, when using the four retrieval configurations (Table 6.4), the mid-infrared retrievals within this section only use 16 channels, and the MIR+FIR retrievals use all 26 channels shown in Figure 6.10.

The first set of retrievals have been performed on the LBLDIS GHM simulation described

in Section 6.4.1, as the minimisation process in Bantges et al. (2020) found this to be the best fit simulation to the observation examined in this chapter. These retrievals are used to assess the impact of the fast scattering approximations used within RTTOVv12 (Section 6.5.1). The second set of retrievals have been then performed on the FORUM-aircraft observation, where the ability of the bulk optical property models will influence the fitting process (Section 6.5.2).

6.5.1 Retrieval from the simulation

As described in Section 6.2, RTTOVv12 uses the Chou approximation to simulate cloudy radiances. This approximation introduces a positive bias into the spectrum within the far-infrared that is dependent on the cloud parameters (Martinazzo et al. 2021). LBLDIS, however, as described in Section 2.1.3 and shown in Section 6.4.1, uses numerical methods to calculate the ice cloud interactions. Therefore, by performing a retrieval on the LBLDIS GHM FORUM-aircraft simulation described in Section 6.4.1, it can be determined if the Chou approximation significantly hinders the retrieval capability as the inputs are known. The simulated spectrum has had Gaussian noise applied based on the FORUM-aircraft uncertainty (grey in Figure 6.8), which is the combined instrument noise, calibration uncertainty, and the apodisation uncertainty (shown individually in Figure 6.3b).

Figure 6.12a shows the BT residuals associated with the retrieval from the FORUM-aircraft simulation with the four different retrieval configurations. When the correct (GHM) bulk optical property model and all 26 cloud-sensitive channels are used, the retrieved BT between $400\text{-}500\text{ cm}^{-1}$ is, on average, within 0.2 K of the input simulation. Furthermore, the retrieved values in Table 6.5 capture the input parameters within two ESD. This suggests that despite the Chou approximation's bias in the far-infrared, the retrieval scheme can reasonably estimate true values.

All four retrieval configurations also overestimate the BT in the channel at $\sim 730\text{ cm}^{-1}$. This channel was selected due to its sensitivity to CER despite its increased uncertainty from the FORUM-aircraft apodisation (Figure 3.6). The weaker fitting in this channel has likely contributed towards the larger CER ESD in Table 6.5, however, the input CER is still captured within one ESD for all the retrievals.

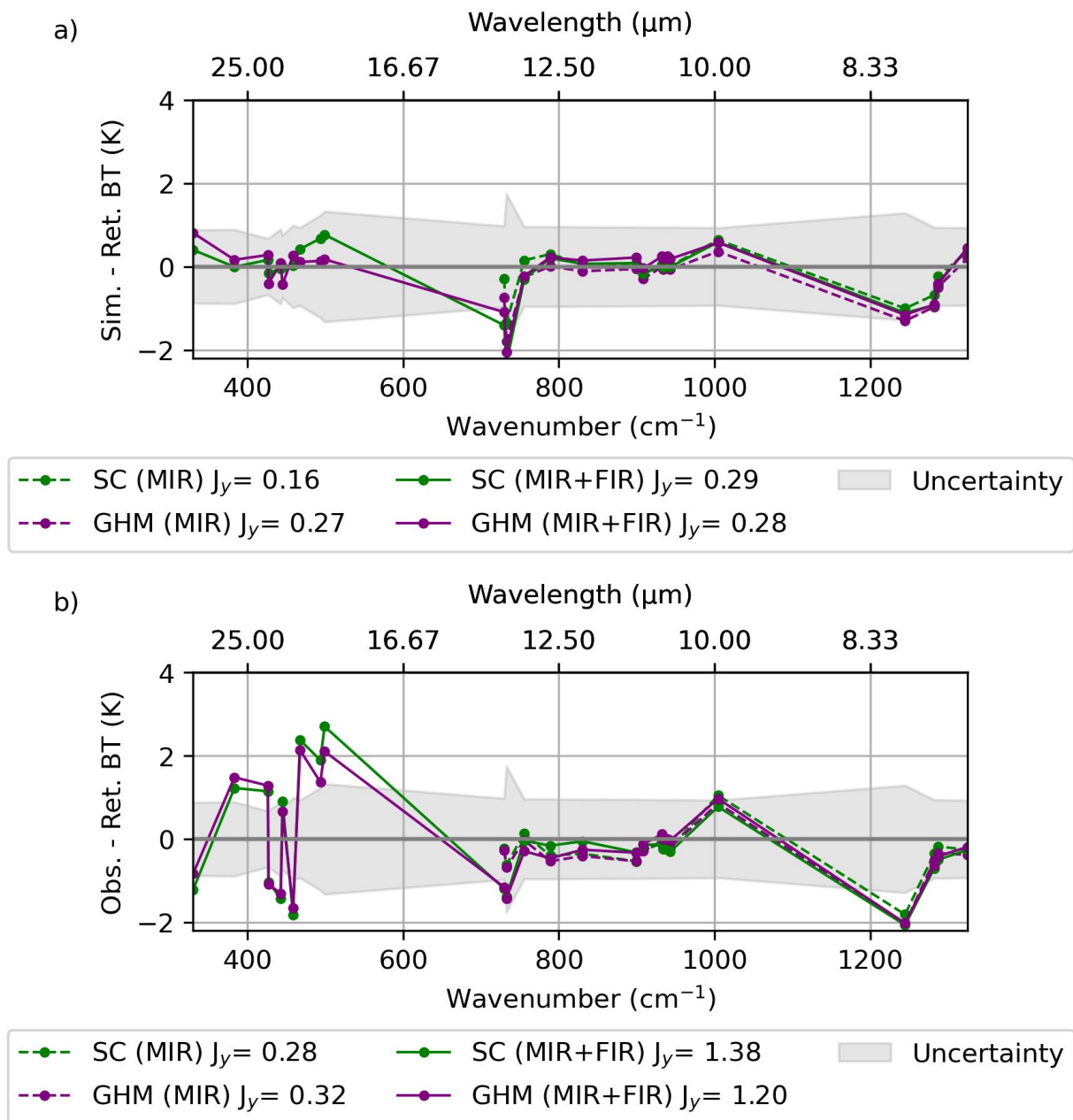


Figure 6.12: The BT residuals between the observed and retrieved spectrum and J_y in the cloud-sensitive channels using the (dashed) mid-infrared (MIR) only and (solid) mid- and far-infrared (MIR+FIR) on the FORUM-aircraft (a) LBLDIS simulation and (b) observation. The residuals within the mid-infrared are partially obscured by the combined MIR+FIR retrieval. The retrievals using the SC and GHM models are shown in green and purple, respectively. The square-root-diagonal of S_y is shaded in grey as the uncertainty.

Table 6.5: The retrieved COT, CER, and CTH from the LBLDIS GHM FORUM-aircraft simulation described in Section 6.5.1 in comparison to the input values for the simulation. Uncertainties shown are one ESD. J_y is also shown for each retrieval configuration and is the same as in Figure 6.12a.

	COT	CER (μm)	CTH (km)	J_y
Input/True State	0.82	34	9.0	-
MIR (GHM)	0.78 ± 0.03	41 ± 9	10.2 ± 2.7	0.27
MIR (SC)	0.74 ± 0.04	40 ± 9	10.7 ± 2.3	0.16
MIR+FIR (GHM)	0.84 ± 0.04	38 ± 8	9.5 ± 0.3	0.28
MIR+FIR (SC)	0.82 ± 0.04	35 ± 8	9.1 ± 0.3	0.29

Given this, the impact of including the far-infrared in retrievals from the simulation can now be examined. Using the mid-infrared alone suggests that the SC model is a better representation of the ice crystal habit mix, with a measurement cost 40 % smaller than with the GHM model (Table 6.5). This is incorrect as the GHM model was used in the simulation. When the far-infrared is included, the residual associated with the GHM model is up to 0.6 K smaller than its counterpart between $400\text{--}450\text{ cm}^{-1}$ (Figure 6.12a) comparable to expected residuals for CER in this range (Figure 6.11). This is indicative of a better fit, and correctly suggests the GHM model is the better representation of the ice crystals within the simulation. However, this improvement is not significantly reflected in the measurement cost, with only a slightly smaller J_y for the GHM model. This is likely due to the larger BT residual for the GHM than SC habit in the $\sim 730\text{ cm}^{-1}$ channel countering the reduced residual in the far-infrared. More generally, including the far-infrared leads to a closer agreement between all of the retrieved values and the known truth (Table 6.5), as well as a significant reduction in the uncertainty associated with the CTH.

6.5.2 Initial retrieval from the observation

As in the previous section, retrievals of COT, CER, CTH, and ice crystal habit have been performed with a fixed vertical profile, but now on the real FORUM-aircraft radiance observation. Given that the fixed vertical profile is only an approximation of the true state, all the retrieval targets will not be explored in detail as these will be discussed in the context of the

full retrieval in Section 6.6. In this section, only the retrieved ice crystal habit and BT residuals are discussed to help separate the influences of the vertical profile and cloud approximations on the full retrieval in Section 6.6 Di Natale & Palchetti (2022), Di Natale et al. (2024).

The in-situ measurements of the ice crystal habits suggest that the GHM model is a better representation of the ice crystals within the cloud than the SC model (Section 6.3.1). Figure 6.12b shows the BT residuals for retrievals from the FORUM-aircraft observation using only the cloud-sensitive channels and similar trends to the simulated case are observed. The measurement cost per channel using the mid-infrared alone is again slightly larger for the GHM model than the SC model, with retrieved spectra within 0.2 K of each other. When far-infrared channels are introduced, J_y is reduced by 13 % using the GHM model. This reduction is driven by the fitting between 450-500 cm^{-1} , where there are differences of up to 0.6 K between the MIR+FIR SC and GHM retrievals. In common with the simulated results from Section 6.5.1, this strongly implies that for this case study, observations from the far-infrared are needed to correctly diagnose the ice crystal habit.

However, the BT residuals within the far-infrared generally exceed the measurement uncertainty, particularly between 450-500 cm^{-1} , while there is little change in the mid-infrared residuals with and without the far-infrared. This reaffirms the findings in Bantges et al. (2020), which highlighted the inability of current bulk optical scattering models to simultaneously match radiance signals in the mid- and far-infrared. Despite this, the bulk optical property models do not prevent the inclusion of the far-infrared from distinguishing between ice crystal habits.

It is important to reiterate that the temperature and water vapour profile are currently fixed to measurements from the dropsonde. To test that the fixed temperature and water vapour profile is not biasing the habit determination, the simultaneous retrievals of temperature and water vapour from the radiance observation are now included in the next section.

6.6 Retrieval of Temperature, Water Vapour, and Cloud Properties

Retrievals of temperature, water vapour, COT, CER, CTH, and ice crystal habit have been performed using both the clear-sky and cloud-sensitive channels shown in Figure 6.13 on the FORUM-aircraft observation. The same four configurations described in Table 6.4 are used, however the mid-infrared retrievals now use 116 channels, and the MIR+FIR retrievals use 226 channels (Figure 6.10). In each case, the a-priori and a-priori covariance for temperature, water vapour, COT, CER, and CTH are as described in Section 6.4.3.

The following discussion has been divided into three subsections: Section 6.6.1 focuses on the retrieval of the ice crystal habit, Section 6.6.2 explores the influence of ice crystal habit and the far-infrared on the retrieved temperature and water vapour profiles, before Section 6.6.3 evaluates the retrieved cloud properties against the in-situ measurements described in Section 6.3.1.

6.6.1 Retrieval of the Ice Crystal Habit

As discussed in Section 6.3.1, the in-situ observations of the ice crystals suggest that the GHM model is a better representation of the ice crystals within the cloud. Figure 6.13 shows that using the mid-infrared alone does not allow a clear distinction between the GHM and SC models when only the spectral fit is considered. In the cloud-sensitive channels, comparable values of J_y for the MIR GHM and MIR SC configurations are seen, similar to the findings in Section 6.5.2. This is also true across all 116 mid-infrared channels which include greater spectral sensitivities to the temperature and water vapour vertical profile. This suggests that while the inclusion of temperature and water vapour in the retrieval does affect the BT residuals, it does not significantly impact the retrieval's inability to distinguish between habits when only the mid-infrared is used.

When the far-infrared is included, there is a clearer distinction between habit models than when only the mid-infrared channels are used. This is shown by the J_y for the MIR+FIR GHM configuration being 9 % smaller than its SC counterpart in the cloud-sensitive channels, while

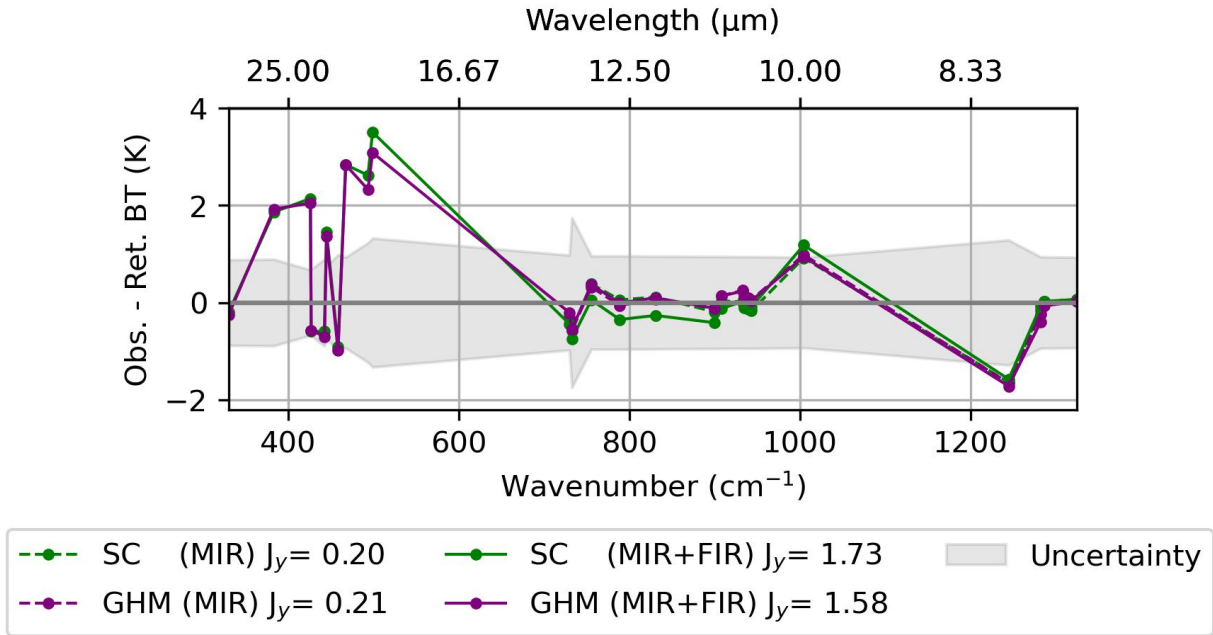


Figure 6.13: The BT residuals between the observed and retrieved spectrum and J_y for retrievals from the observation. All 226 channels are used in the retrieval, but only residuals and J_y in the 26 cloud sensitive channels are shown for comparison to Figure 6.12. The square-root-diagonal of S_y is shaded in grey as the uncertainty. J_y across all 116 (MIR) and 226 (MIR+FIR) channels is as follows: SC (MIR): 0.33. GHM (MIR) 0.34. SC (MIR+FIR): 1.20. GHM (MIR+FIR): 1.15.

the MIR equivalents have comparable measurement costs in the cloud-sensitive channels. This smaller J_y for the MIR+FIR GHM configuration suggests that the GHM model enables a better spectral fit, and so is the more likely representation of the ice crystals within the cloud. This is in line with the in-situ measurements.

The addition of temperature and water vapour in the MIR+FIR retrieval has a more notable impact on the spectral distinction between ice crystal habits than when only the mid-infrared channels are used. This is most clearly seen when the BT residuals associated with the MIR+FIR SC and GHM configurations within the far-infrared are compared (Figure 6.13). Below 410 cm^{-1} , the BT residuals between the GHM and SC configurations are now comparable. Between 410 and 600 cm^{-1} the residuals differ by, on average, 0.1 K , which is 0.5 K smaller than when the vertical temperature and water vapour profiles were fixed to the dropsonde measurements (Figure 6.12b). This similarity is reflected by the reduction in percentage difference between J_y in the cloud-sensitive channels for the MIR+FIR GHM and SC configurations (by 6%) relative to the retrievals with the fixed vertical profile.

Furthermore, when J_y across the clear-sky and cloud-sensitive channels is considered, the percentage difference between J_y for the MIR+FIR SC and GHM configurations is only 5 %. This effect is a result of the differences in the associated retrieved temperature and water vapour profiles and will be explored further in Section 6.6.2.

6.6.2 Retrieval of Temperature and Water Vapour

The benefits of including the far-infrared in clear-sky retrievals of upper tropospheric water vapour have been explored using observations in mid-latitude conditions (Warwick et al. 2022, Panditharatne et al. 2025). These results indicate that including the far-infrared increases the DOFS for temperature and water vapour from 2.9 to 3.8 and from 1.9 to 3.7, respectively, due to the additional information contained within the far-infrared spectrum. Both the MIR and MIR+FIR GHM retrievals generally capture the AK-treated dropsonde profile within 2 ESD (not shown). However, as previously discussed (Section 6.6.1), including temperature and water vapour in the retrieval influences the determination of the ice crystal habit for the retrievals that include the far-infrared. This is in part due to the increased sensitivity of far-infrared radiances to the water vapour and temperature profile as reflected in the DOFS.

Figure 6.14 shows the retrieved temperature and water vapour profiles using all 226 channels in the MIR+FIR GHM and SC configurations. The temperature retrieval for both habit models is worse than the a-priori with residuals of up to 1 K from the AK-treated dropsonde temperature profile (Figure 6.14b). This is in part due to the increased uncertainty in the $15 \mu\text{m}$ CO_2 wings following the FORUM-aircraft apodisation (Chapter 3). Despite this, the AK-treated dropsonde temperature profile is within one ESD of both retrievals below 6 km, and entirely within the GHM retrieval. For the MIR+FIR case, assuming the SC habit reduces the retrieved temperature between 2-8.5 km relative to the GHM case, with differences reaching up to 1.9 K. A reduction is also seen between the SC and GHM cases if only the mid-infrared channels are used, but in this case it is smaller, reaching a maximum of 0.4 K (not shown). This reduction in temperature lowers the simulated BT throughout the channels, particularly between $450\text{-}500 \text{ cm}^{-1}$ to balance the changes in the cloud retrieval products that will be discussed in Section 6.6.3.

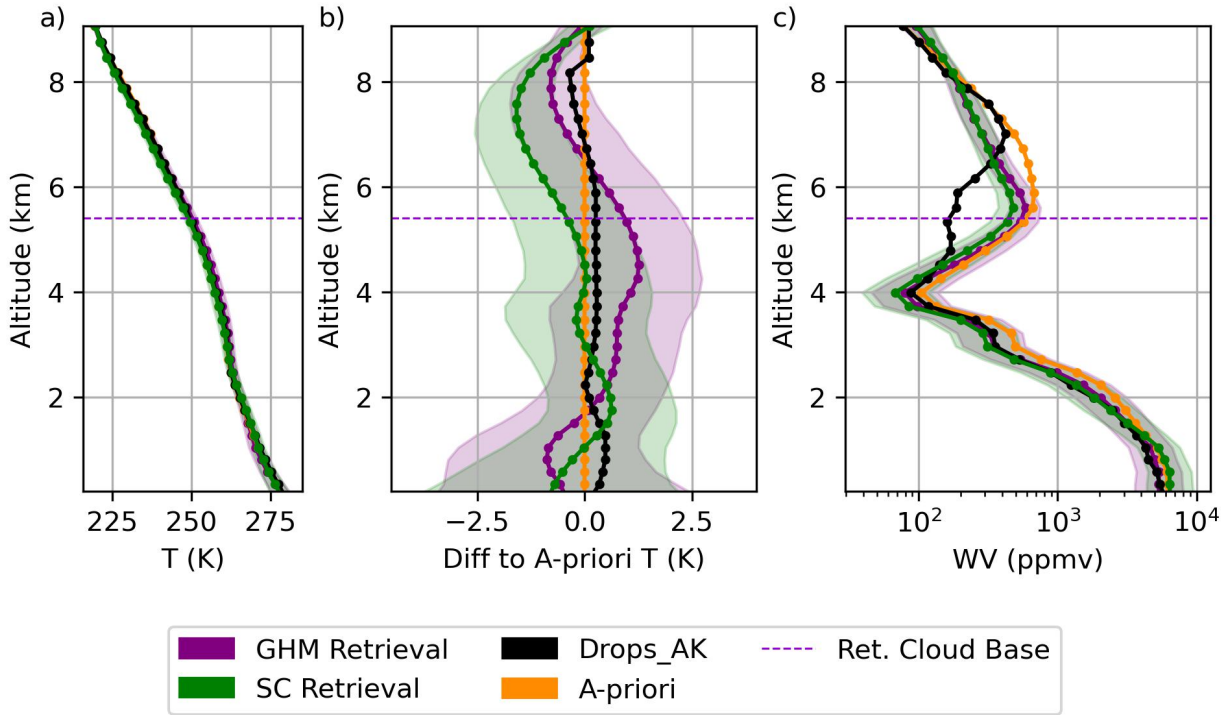


Figure 6.14: The retrieved (a) temperature and (c) water vapour profile from the FORUM-aircraft observation with (b) differences between the retrieved and AK-treated dropsonde temperature profiles. The retrievals assuming the GHM and SC models are shown in purple and green, respectively, and use all 226 channels. The average cloud base from the two retrievals is shown with the dashed purple line. The shaded region around each retrieval represents one ESD. The a-priori is shown in orange, with the surrounding shading showing one standard deviation. The AK-treated dropsonde (Drops_AK) profiles are shown in black. The uncertainty in the dropsonde measurements are discussed in Section 6.3 but are not visible on the scale of this figure. For temperature, the dropsonde uncertainty is 0.2 K. For water vapour, the dropsonde uncertainty varies with altitude but remains below 9%.

Figure 6.14c shows that the habit does not affect the retrieved water profile outside of one ESD, with the median retrieval bias throughout the vertical only increasing by 2 % from the GHM to SC model. Both retrievals capture the AK-treated dropsonde profile below 4 km and above 8 km, but struggle at the base of the cloud. Neither retrieval captures the observed dry patch around the cloud base, as they remain close to the a-priori. This is likely due to a combination of the sensitivity of the water vapour retrieval to the cloud position and dry bias of the dropsondes that was discussed in Section 5.5.2.

6.6.3 Retrieval of COT, CER, and CTH

Figure 6.15 shows the retrieved values for the COT and CER in comparison to the measurements of the cloud described in Section 6.3.1. The retrieved values are plotted at the midpoint of the retrieved cloud to allow a comparison to the individual CERs derived from the PSDs. While the lidar-derived COT was taken 4 s after the radiance observation, the in-situ measurements of the cloud were taken between 10:32 and 11:32 UTC, and so the Level 2 COT and CER from the MODIS Terra overpass at 10:40 UTC are also included. These have been averaged across the spatial range of the in-situ measurements and radiance observation (58.7-59.75°N and 4.2-3.3°W).

There is some evidence of systematic behaviour between the COT retrievals shown in Figure 6.15a. For example, the GHM retrievals (purple points) show slightly larger COTs than their corresponding SC retrievals. Similarly, the MIR retrievals (square points) show slightly larger COTs than the MIR+FIR retrievals. However, all 4 configurations retrieve the COT within the uncertainty of the lidar-derived COT of 0.8 ± 0.1 that was co-located with the radiance observation, and are within one median absolute deviation of the median MODIS COT taken approximately an hour after (Figure 6.15a). The relative insensitivity of the COT retrieval to the inclusion of the far-infrared is expected given the strong sensitivity to COT across most of the mid-infrared channels used in the retrieval (Bantges et al. 2020), and this is reflected in the minimal increase in the DOFS from 0.9 to 1.0 when the far-infrared is included.

The centre of the cloud measured by the lidar is at ~ 7.5 km, and this is captured within the range of all 4 retrieval configurations, with a consistent DOFS of 1 for CTH across all the retrievals. The inclusion of the far-infrared (triangle points) slightly shifts the cloud higher by ~ 0.4 km, but the retrievals still remain within one ESD of their MIR counterparts (square points). In contrast, assuming the SC model rather than the GHM model can be seen to slightly lower the retrieved cloud by ~ 0.4 km, and this is likely to counteract the colder vertical profile that is retrieved with this habit (Figure 6.14).

For both the retrieved CTH and COT, the ESD halves from 0.8 to 0.4 km and 0.06 to 0.03, respectively, with the inclusion of the far-infrared. This suggests a greater confidence in the state and agrees with previous simulation studies that suggest including the far-infrared

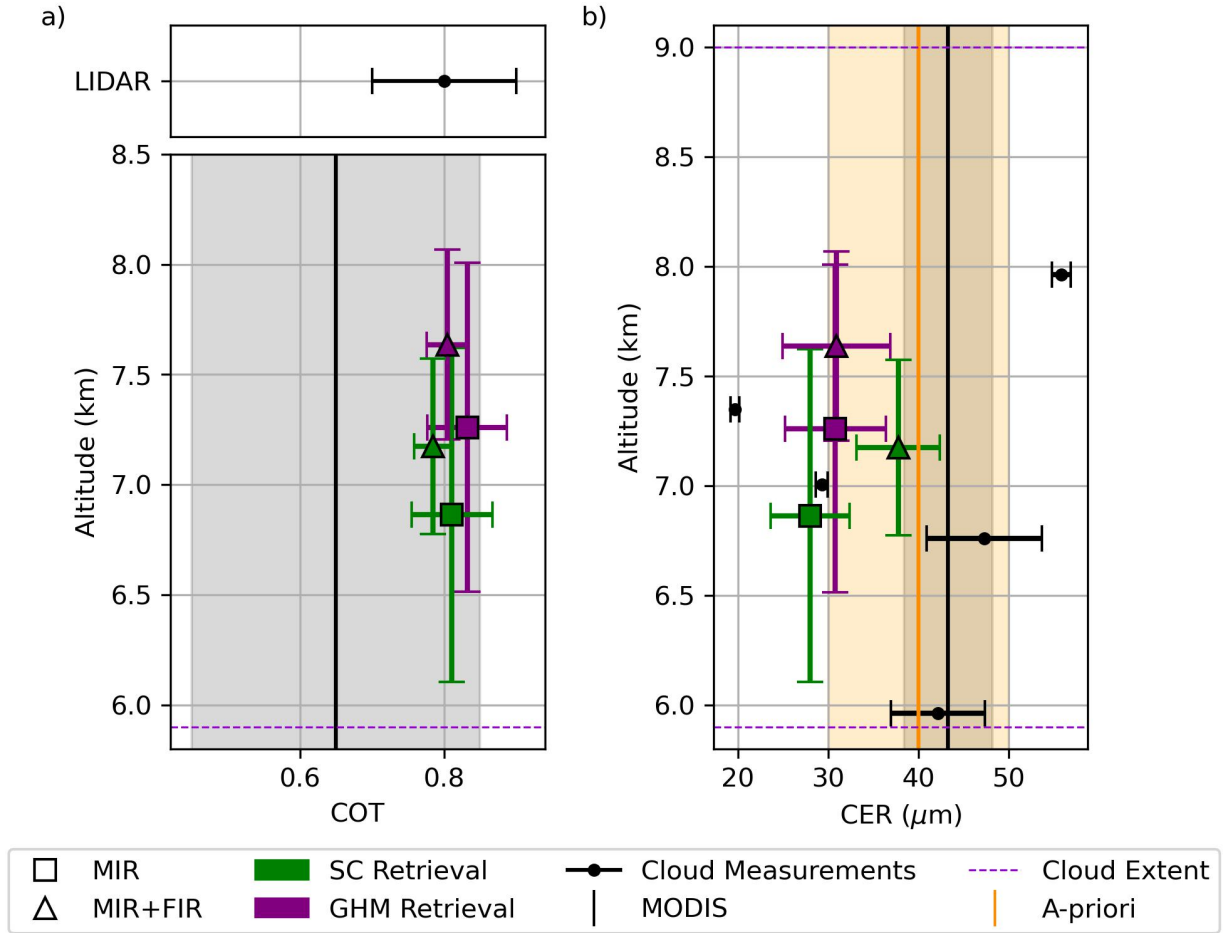


Figure 6.15: The retrieved (a) COT and (b) CER from the FORUM-aircraft observation in comparison to measurements of the cloud. The retrieved values are shown using squares (116 MIR channels) and triangles (226 MIR+FIR channels). The retrievals using the SC and GHM models are shown in green and purple, respectively. Error bars show one ESD in the retrieved CTH, COT, or CER. The measurements of the cloud taken by instruments on board the plane are shown as black dots. For the COT, the mean value derived from the lidar measurements between from 09:48:09 to 09:49:09 UTC is shown, with the error bar representing the standard error of the mean. For the CER, the black dots are calculated from the PSDs measured by the Cloud 2-DS probe at 5 altitudes within the cloud, and the error bars show one standard deviation derived from the bimodal fit. The vertical black line shows the median MODIS Level 2 COT and CER from the 10:40 UTC overpass between $58.7\text{--}59.75^\circ\text{N}$ and $4.2\text{--}3.3^\circ\text{W}$ at 1 km resolution. The shading around the MODIS COT and CER is one median absolute deviation and shows the spread of values within this spatial region. The a-priori for the CER is shown in orange with shading representing one standard deviation. The a-priori for the COT is 0.01 and is not shown.

could reduce uncertainties in cloud retrieval targets (Libois & Blanchet 2017).

All 4 retrievals of the CER are between 28 and $38 \mu\text{m}$. This is generally lower than the a-priori and MODIS CER, and tends towards the smaller in-situ measurements (Table 6.3). There is a notable difference in sensitivity to the inclusion of far-infrared information

depending on the habit selected in the retrieval. For the GHM case, the retrieved CER is unchanged at $31 \pm 6 \mu\text{m}$ regardless of whether far-infrared channels are included, showing good agreement with the weighted mean of the in-situ CER measurements ($32 \pm 14 \mu\text{m}$). In contrast, if the SC habit is selected, the retrieved CER increases from 28 ± 4 to $38 \pm 5 \mu\text{m}$ from the MIR only to the MIR+FIR case, with the MIR+FIR SC retrieval remaining closest to the a-priori. There is also a slight increase in the CER DOFS from 0.7 to 1.0 when the far-infrared is included.

Overall, despite the limitations associated with the bulk optical property models, and the use of a fast cloud scattering approximation, the retrieved cloud properties agree with the independent lidar and in-situ observations. The addition of far-infrared information provides a greater constraint on the inferred COT and CTH (reduced retrieval uncertainty) and, importantly, allows improved discrimination of crystal habit compared to retrievals using only the mid-infrared part of the spectrum.

6.7 Conclusions

This chapter presents the first retrievals of cirrus cloud properties (optical thickness, effective radius, cloud position, habit), temperature, and water vapour that exploit upwelling mid- and far-infrared radiances ($100\text{--}1600 \text{ cm}^{-1}$) observed from an aircraft. These airborne radiances have been adapted to emulate the FORUM Sounding Instrument's spectral characteristics to provide an indication of the benefit that the FORUM satellite mission could bring to our current understanding of ice clouds.

Additional coefficients have been built to enable IMS to perform retrievals assuming different ice crystal habits. Despite the use of the Chou approximation to simulate cloudy radiances, sensitivity tests showed that it does not compromise the ability of IMS to retrieve the 'true' state for this case.

The primary aim of this study was to investigate whether the inclusion of the far-infrared improved the quality of the retrieved targets, with a particular focus on the ice crystal habit. To do this, four different retrievals were performed on the observation: with and without the

far-infrared, and with either the Solid Column or General Habit Mix bulk optical property model (Yang et al. 2013, Baum et al. 2014). The outputs were then evaluated against co-located measurements from the onboard lidar and in-situ measurements of the ice crystal habits and particle size distributions that were taken an hour after the radiance observation.

The retrieved cloud optical depth and position of the cloud were consistent with the lidar measurements for all four configurations. However, the inclusion of the far-infrared halved the retrieval uncertainties for these retrieval targets in comparison to when only the mid-infrared was used. These results are in line with previous findings based on theoretical simulations and ground-based observations (Libois & Blanchet 2017, Di Natale et al. 2017).

These results also demonstrate that including the far-infrared enabled a clearer and more likely distinction of the ice crystal habit for this case. When only the mid-infrared was used, the spectral fit suggested that the Solid Column model was a slightly better representation of the ice crystals within the cloud, which conflicted with the in-situ measurements. The addition of the far-infrared resulted in a better spectral fit for the General Habit Mix, which was more compatible with the habit distribution inferred from measurements. Furthermore, the choice of habit had more of a significant influence in the retrieval of both temperature and the cloud effective radius when the far-infrared was included.

It is important to note that despite these encouraging results, there are caveats associated with this study. The in-situ measurements that were used to derive the cloud effective radius were taken an hour after the radiance measurements, and it is possible they may have evolved with time (O'Shea et al. 2016). Perhaps more critically, the spectral residuals obtained after the retrievals have converged exceed the measurement uncertainty within the far-infrared. While these results imply that this does not preclude using the selected (and widely used) Yang et al. (2013) and Baum et al. (2014) ice optical property models chosen here, it does imply that they need refinement to be consistent across the entire infrared (Bantges et al. 2020). This finding is in full agreement with previous work and motivates both the further development of these models, and the need for a larger set of coherent observations of in-situ cloud properties and spectral radiances across the entire infrared to enable more comprehensive tests across a range of cirrus types.

Chapter 7

Conclusions and Future Work

This thesis has presented the extension of the Rutherford Appleton Laboratory Infrared Microwave Sounding (IMS) retrieval scheme into the far-infrared in preparation for the European Space Agency's Far-infrared Outgoing Radiation Understanding and Monitoring (FORUM) mission. This retrieval scheme has been optimised to exploit far-infrared signals of water vapour and ice clouds, and it has been applied to observations of upwelling far-infrared radiances that have been adapted to mimic the FORUM Sounding Instrument's (FSI) spectral characteristics.

The clear-sky work in Chapters 3, 4, and 5 has been published in Panditharatne et al. (2025). The work associated with ice clouds in Chapters 3 and 6 has been submitted in Panditharatne et al. (under review).

Chapter 3 details the method used to create the FORUM-aircraft observations that are used throughout this thesis. These observations are based on coincident measurements of upwelling mid- and far-infrared radiances taken by the Tropospheric Airborne Fourier Transform Spectrometer (TAFTS) and the Airborne Research Interferometer Evaluation System (ARIES) during two different aircraft campaigns. The observations from each instrument have been combined and modified to mimic the FSI's spectral characteristics to approximate the spectral information that observations taken by FSI could have. This process increased the uncertainty in the measured mid-infrared radiances, particularly in the $15\text{ }\mu\text{m}$ CO_2 absorption band. However, testing on simulations of cloudy radiances suggested that these effects are partially reduced by the presence of clouds.

In Chapter 4, the IMS retrieval scheme was optimised for clear-sky retrievals of FSI observations. Three key modifications were introduced into the scheme to build the new FSI configuration. First, the ability of the forward model Radiative Transfer for the TIROS Operational Vertical Sounder v12 (RTTOVv12) was tested in simulating FSI radiances. Second, the spectrally dependent covariance matrices used to constrain the retrieval were configured for the FSI. The measurement covariance was based on the FSI's expected apodised noise-equivalent-spectral-radiance (NESR) and target absolute radiometric accuracy (ARA), and the a-priori covariance for surface spectral emissivity was extended into the far-infrared using the Huang et al. (2016) surface emissivity database. Third, an information content analysis was performed to optimise the channels used in the retrieval, resulting in the selection of 200 channels for temperature and water vapour retrievals. The highest channel density was seen in the $400\text{--}600\text{ cm}^{-1}$ region due to the sensitivity of radiances to water vapour and temperature coupled with the relatively low NESR and ARA in this region.

The performance of IMS was assessed at each iteration and compared to IMS's default Infrared Atmospheric Sounding Interferometer (IASI) configuration. The goal of this comparison was not to determine which instrument resulted in a better retrieval but rather to check the new, extended IMS code. This assessment was done using 240 RTTOVv12 simulations of top-of-atmosphere (TOA) upwelling radiances based on MERRA-2 reanalysis profiles. Retrievals using the FSI configuration had a tighter measurement covariance than the IASI configuration below $\sim 1200\text{ cm}^{-1}$. Both configurations demonstrated comparable retrieval biases. However, slight improvements in the FSI median retrieval bias were observed in the mid- to upper troposphere for water vapour, with a reduced median uncertainty of at most 7 %, and an increase of ~ 1 degrees of freedom for signal for both water vapour and temperature due to additional information in the upper troposphere. It is worth noting that in this comparison, the measurement covariance used for IASI is not the quoted instrument NESR and ARA but instead implicitly includes the effects of instrumental and forward model error. For the FSI, the instrumental error has been approximated using the target instrument specifications, so it is likely that the comparison overstates the improvement that FORUM will bring relative to IASI observations.

Chapter 5 describes the testing of IMS on coincident aircraft observations of mid- and far-infrared upwelling radiances taken in clear-sky conditions. These were taken during the C153 flight as a part of the PIKNMIX-F campaign, which included periodic releases of dropsondes to measure the atmospheric state. Eight observations closest to each dropsonde release were selected and modified to create FORUM-aircraft observations. Based on the uncertainty associated with these observations, a FORUM-aircraft configuration was developed within IMS. This only differed from the FSI configuration in the measurement covariance (now based on the FORUM-aircraft measurement and apodisation uncertainty) and channel selection used for the retrieval. The increased uncertainty in the $15\text{ }\mu\text{m}$ CO_2 band wings propagated through to the selection of channels used in the retrieval. However, applying this revised FORUM-aircraft configuration to 240 simulated aircraft-level radiances based on the MERRA-2 test set showed only minimal differences in the retrieved temperature and water vapour profiles compared to the retrievals using the FSI configuration from the equivalent TOA radiances. This gives confidence that the results obtained here from the aircraft observations provide a good indication of what might be expected from FORUM when it is operating.

Retrievals of the FORUM-aircraft observations required the a-priori covariance for the temperature to be tightened to the local ERA5 grid of the flight as a result of the larger measurement uncertainty associated with the $15\text{ }\mu\text{m}$ CO_2 band. This enabled simultaneous temperature and water vapour retrievals exploiting the full FSI spectral range. Tightening this constraint had the additional benefit of reducing the bias in the water vapour retrieval by up to 33 %. The final retrievals of temperature and water vapour generally captured the dropsonde measurements of the atmospheric state, with the Root-Mean-Squared-Error (RMSE) values in water vapour concentration reducing by 18 % from the a-priori to the retrieved state across all eight cases. However, these results do still highlight some inconsistencies, with both the ERA5 a-priori and retrievals consistently overestimating the humidity between 5 and 7 km and a persistent cold bias below 2 km in seven out of the eight cases analysed. These deficiencies are manifested in the radiance residuals seen in the final fitted spectra, and suggest that the humidity bias may be related to water vapour spectroscopic uncertainty in the far-infrared. However, these differences may also be caused by a dry bias observed in

the model of dropsondes used in this work. While this analysis implies that the variability around each dropsonde is relatively small, the near-surface bias may be partly related to the dropsonde drift away from the aircraft as it descends through the atmosphere.

The IMS scheme has now been extended to cover the far-infrared spectral range in preparation for the FORUM mission. It has been tested on multiple aircraft-level observations of upwelling far-infrared radiances and a diverse set of TOA-simulated upwelling radiances, exploiting channels from the full FSI spectral range. Pending the real FSI systematic and random uncertainties, IMS is ready to use for clear-sky retrievals from the FSI observations.

Chapter 6 presents the first retrievals of cirrus cloud properties (optical thickness, effective radius, cloud position, habit), temperature, and water vapour that exploit upwelling mid- and far-infrared radiances observed from an aircraft. These airborne radiances were taken as a part of the B895 flight during the CIRCCREX campaign on 13 March 2015, and have again been adapted to emulate the FSI's spectral characteristics.

Before this work, IMS could retrieve the optical thickness, effective radius, and position of the cloud but could not perform retrievals of the ice crystal habit. Additional coefficients have been built to enable IMS to perform these retrievals by assuming different ice crystal habits based on the Yang et al. (2013) and Baum et al. (2014) cloud optical property models (General Habit Mix (GHM), Aggregate Solid Columns (ASC), and Solid Columns (SC)). The use of the Chou approximation within RTTOVv12 reduces the spectral residuals between far-infrared radiances simulated using the GHM and ASC models to below the FORUM-aircraft instrument uncertainty. However, the spectral residuals between GHM and SC simulated radiances still exceeded the FORUM-aircraft measurement uncertainty, and so, in principle, the different habits can be distinguished when performing retrievals of the aircraft observations. Sensitivity tests on simulations based on the B895 flight also showed that using the Chou approximation did not compromise the ability of IMS to retrieve the 'true' state for this case.

The primary aim of the study was to investigate whether the inclusion of the far-infrared improved the quality of the retrieved targets, with a particular focus on the ice crystal habit. To do this, four different retrievals were performed on the observation: with and without the

far-infrared and with either the SC or GHM optical property models. Twenty-six additional channels were included to improve the retrievals of ice cloud properties. These channels were selected based on the spectral sensitivities to the optical thickness, effective radius, and habit. The retrieved properties were then evaluated against co-located measurements from the onboard lidar and in-situ measurements of the ice crystal habits and particle size distributions that were taken an hour after the radiance observation. The measurements of the particle size distributions were used to derive the cloud effective radius at different altitudes within the cloud.

The retrieved cloud optical depth and position of the cloud were consistent with the lidar measurements for all four configurations. However, including the far-infrared halved the retrieval uncertainties for these targets compared to when only the mid-infrared was used. These results align with previous findings based on theoretical simulations and ground-based observations (Libois & Blanchet 2017, Di Natale et al. 2017).

These results also demonstrate that including the far-infrared enabled a clearer distinction of the ice crystal habit for this case. When only the mid-infrared was used, the spectral fit suggested that the SC model slightly better represented the ice crystals within the cloud, which conflicted with the in-situ measurements. The addition of the far-infrared resulted in a better spectral fit for the GHM model, which was more compatible with the in-situ measurements of the ice crystal habits. Furthermore, the choice of habit had more of a significant influence on the retrieval of both temperature and the cloud effective radius when the far-infrared was included.

It is important to note that despite these encouraging results, there are caveats associated with this study. The in-situ measurements that were used to derive the cloud effective radius were taken an hour after the radiance measurements and it is possible they may have evolved over time. Perhaps more critically, the spectral residuals obtained after the retrievals have converged exceed the measurement uncertainty within the far-infrared. While these results imply that this does not preclude using the selected (and widely used) optical property models chosen here, it does imply that they need refinement to be consistent across the entire infrared. Nonetheless, the work in Chapter 6 provides the first observational confirmation

from upwelling radiances measured above cirrus that the far-infrared will allow an improved discrimination of crystal habit and reduced uncertainty in optical depth and cloud top height. These parameters are absolutely critical in the context of better constraining both the current net radiative effect of cirrus and the magnitude and sign of future cirrus feedback.

7.1 Suggestions for Future Research

There are multiple streams of research that could follow from the work presented in this thesis. One of the most significant streams follows the implications of an improved understanding of water vapour distributions and ice cloud properties that comes with retrievals exploiting the far-infrared. Once the FORUM satellite has launched, a better knowledge of these research areas could be manifested in more accurate representations within climate or radiative transfer models. In preparation for the FORUM mission, there are 4 priority topics described here that are linked to the work in this thesis and deserve further exploration: (i) support for a calibration and validation (Cal/Val) campaign; (ii) implementing better ice crystal representations; (iii) retrievals from downwelling observations; (iv) new observations within the far-infrared.

Calibration/Validation Campaigns

The large number of FORUM-aircraft observations available during the C153 flight presents an opportunity to investigate how significant the water vapour variability within the scene could be for a Cal/Val campaign for the FORUM mission. Cal/Val campaigns investigate the calibration uncertainty of satellite-borne instruments, in this case the FSI, to validate the quality of measured radiances and retrieved products. One method is to use an airborne instrument to underfly the satellite so that they simultaneously measure the same scene. To fairly compare observations from the airborne and satellite instruments, it is useful for the atmosphere to be well characterised, for both instruments to have similar spectral characteristics, and for the scene to be homogeneous. Scene homogeneity can be most easily achieved by limiting analysis to clear-sky conditions (Chander et al. 2013, Newman et al.

2012, Niro et al. 2021).

All three of these requirements are satisfied by the observations in the C153 flight through the creation of the FORUM-aircraft observations, the dropsondes released throughout the flight, and the clear-sky conditions. During these Cal/Val campaigns, it is advantageous to average a large number of aircraft observations within the satellite's ground footprint to reduce the uncertainty associated with the airborne instrument's noise (Newman et al. 2012, Naethe et al. 2023). However, this averaging depends on the assumption that the scene is homogenous. Observations from the C153 flight could be used to investigate a desirable variability limit during Cal/Val campaigns, particularly given the strong sensitivity of water vapour to the far-infrared.

Ice Crystal Representation

There is currently work exploring the inclusion of the temperature-dependence of the refractive index of ice within cloud optical property models (Wang et al. 2024). The implications of these adjustments on the retrieved ice cloud properties could be further investigated. Furthermore, the cloud optical property models and coefficients built for RTTOVv12 are currently defined for specific habits, resulting in the discrete retrievals of ice crystal habit within this thesis. Based on work in Di Natale et al. (2024), a database of coefficients could be constructed that contain continuous fractions of different ice crystal habits. This would enable IMS to use the optimal estimation method to retrieve ice crystal habit instead.

Existing Far-Infrared Observations

The Far-INfrarEd Spectrometer for Surface Emissivity (FINESSE) has recently taken part in two campaigns that measured the downwelling mid- and far-infrared radiances in both clear-sky and cloudy conditions, with in-situ measurements of the atmosphere. The Mixed Phase Clouds and Climate campaign took place in 2022 in Andøya, Norway, and the W-band, HiSRAMS, AERI, FIRR2, FINESSE and FIRMOS Experiment on Remote Sensing campaign took place in 2025 near Montreal, Canada. Currently, a retrieval scheme has not been built for use on the FINESSE observations taken during both of these campaigns. Therefore, there

is potential to adapt the forward model within IMS to simulate downwelling radiances, enabling optimal estimation retrievals of water vapour and ice clouds and potentially surface emissivity from FINESSE observations using IMS.

New Far-Infrared Observations

Currently, there are a limited number of observations of upwelling far-infrared radiances that could be used to improve our understanding of water vapour spectroscopy and cloud optical property models in the far-infrared. The recent launch of the Polar Radiant Energy in the Far Infrared Experiment (PREFIRE) mission offers another opportunity to apply this retrieval scheme to far-infrared radiances. However, its coarser spectral resolution will limit its ability to detect the finer spectral features caused by ice clouds and water vapour absorption (L'Ecuyer et al. 2021).

The TAFTS instrument, which provided the far-infrared observations in this work, was decommissioned in 2022. It is being replaced by the UNiversal InfraRed Airborne Spectrometer (UNIRAS), which is currently under construction and due to be mounted on the Facility for Atmospheric Airborne Measurements (FAAM) aircraft. This instrument includes a 'FORUM-like' and 'IASI-like' configuration that aims to have noise levels comparable to those of their respective satellite-based instruments. This would enable a fairer evaluation of the advantages associated with the FSI in retrievals and more vigorous testing of this retrieval scheme in different atmospheres. Work is currently underway within the FORUM Mission Advisory Group to define future campaigns with UNIRAS. The tool built in this thesis will be applied to exploit future measurements from these scientific studies.

Appendix A: Glossaries of key symbols

This appendix contains two glossaries summarising the key symbols described in Chapter 2.

Table A1: Glossary of key symbols described Section 2.1.

Symbol	Definition
I	Radiance
ρ	Density of a medium
σ_e	Mass extinction cross section
j	Source function coefficient
J	Source function
z	Linear distances normal to the plane (i.e height or pressure axis)
ϕ	Azimuthal angle of the radiation's path
θ	Zenith angle of the radiation's path
μ	$\cos(\theta)$
τ	Optical depth
τ_{surf}	Optical depth at the surface
$\mathcal{T}$	Transmittance
ϵ	Surface spectral emissivity
$B(T)$	Planck function
κ	Imaginary component of the complex refractive index
λ	Wavelength
ω_0	Bulk Scattering Albedo
β_a	Bulk Absorption Coefficient
β_s	Bulk Scattering Coefficient
β_e	Bulk Extinction Coefficient
$P(\mu)$	Phase Function
$\overline{P}(\mu, \mu_i)$	Azimuthally-Averaged Phase Function

Table A2: Glossary of key symbols described Section 2.2.

Symbol	Definition
$\mathbf{x}$	True state vector
$\mathbf{x}_0$	First guess state vector
$\mathbf{x}_a$	A-priori state vector
$\mathbf{x}_{AK}$	AK-treated true state vector that has been smoothed to the vertical resolution and a-priori of the retrieval framework using $\mathbf{A}$
$\hat{\mathbf{x}}$	Retrieved state vector
$\mathbf{S}_x$	Retrieved state covariance
$\mathbf{S}_a$	A-priori covariance
$\mathbf{y}$	Measurement vector
$\mathbf{S}_y$	Measurement covariance
$\mathbf{f}(\mathbf{x})$	Forward function that connects the measurement and state vector
$\mathbf{F}(\mathbf{x}, \mathbf{b})$	Forward model that is used to approximate the forward function
$\mathbf{b}$	Known quantities that influence the forward function that are included in the forward model
$\mathbf{J}$	Cost function composed of the state and measurement cost
$\mathbf{J}_x$	State cost that represents how far the retrieved state has deviated from the a-priori in relation to $\mathbf{S}_x$
$\mathbf{J}_y$	Measurement cost that represents the fit of the forward model output to $\mathbf{y}$ within the limits of $\mathbf{S}_y$
$\mathbf{K}_b$	A matrix of derivatives of the forward model output (radiance) with respect to $\mathbf{b}$
$\mathbf{K}_x$	A matrix of derivatives of the (radiance) with respect to each retrieval product
$\mathbf{A}$	The averaging kernel which shows the sensitivity of the retrieved to the true state
$\mathbf{D}_{F,n}$	Degrees of freedom for noise
$\mathbf{D}_{F,s}$	Degrees of freedom for signal
$\mathbf{G}$	The gain matrix which shows the sensitivity of the retrieval to the measurement and its error
$\mathbf{H}$	Information content showing the factor by which knowledge of a quantity is improved by making a measurement

Appendix B: Water vapour conversions

The water vapour profiles measured by Vaisala RD94 dropsondes are provided in relative humidity (RH) and mass mixing ratio, r . In this thesis, all the analyses are performed using volume mixing ratio (VMR). The conversions presented here are based on descriptions in Salby (1996). The VMR can be calculated from r using the following equation:

$$\text{VMR} = 10^6 r_s \frac{m_{\text{dry air}}}{m_{\text{water}}} \quad (1)$$

where the VMR is in units of ppmv, and r_s is in units of gkg^{-1} .

The uncertainties associated with the dropsonde water vapour measurements have been defined in Section 5.2.1 in relative humidity. These uncertainties are translated into VMR by finding the maximum and minimum RH values and converting them into VMR. To perform this conversion, the saturation vapour pressure, e_s , and saturation specific humidity, q_s , are calculated using Eq. 2 and 3:

$$e_s = 611.12 e^{\frac{l_v}{R_v} \left(\frac{1}{T_0} - \frac{1}{T_d} \right)} \quad (2)$$

$$q_s = \frac{0.622 e_s}{p - 0.378 e_s} \quad (3)$$

where l_v is the latent heat of vaporisation of water, R_v is the specific gas constant of water vapour, T is a reference temperature of 273.15 K, T_d is the dewpoint temperature that is provided by the dropsonde, and p is the pressure measured by the dropsonde.

The saturation mass mixing ratio, r_s , can then be defined using Eq 4:

$$r_s = \frac{q_s}{1 - q_s} \quad (4)$$

and used to calculate the mass mixing ratio in units of kg kg^{-1} :

$$r = \text{RH} r_s \quad (5)$$

This can then be converted into VMR using Eq. 1.

Bibliography

Allan, R. P. (2011), 'The role of water vapour in Earth's energy flows', *Surveys in Geophysics* **33**(3–4), 557–564.

URL: <http://dx.doi.org/10.1007/s10712-011-9157-8>

Allan, R. P., Willett, K. M., John, V. O. & Trent, T. (2022), 'Global changes in water vapor 1979–2020', *Journal of Geophysical Research: Atmospheres* **127**(e2022JD036728).

URL: <http://doi.org/10.1029/2022JD036728>

Andrews, D. G. (2010), *An introduction to atmospheric physics*, 2 edn, Cambridge University Press.

URL: <https://doi.org/10.1017/CBO9780511800788>

Arshad, M., Ma, X., Yin, J., Ullah, W., Liu, M. & Ullah, I. (2021), 'Performance evaluation of ERA-5, JRA-55, MERRA-2, and CFS-2 reanalysis datasets, over diverse climate regions of pakistan', *Weather and Climate Extremes* **33**(100373).

URL: <https://doi.org/10.1016/j.wace.2021.100373>

Bantges, R. J., Brindley, H. E., Murray, J. E., Last, A. E., Russell, J. E., Fox, C., Fox, S., Harlow, C., O'shea, S. J., Bower, K. N., Baum, B. A., Yang, P., Oetjen, H. & Pickering, J. C. (2020), 'A test of the ability of current bulk optical models to represent the radiative properties of cirrus cloud across the mid-and far-infrared', *Atmospheric Chemistry and Physics* **20**, 12889–12903.

URL: <https://doi.org/10.5194/acp-20-12889-2020>

Baran, A. & Francis, P. (2004), 'On the radiative properties of cirrus cloud at solar and thermal wavelengths: A test of model consistency using high-resolution airborne radiance

measurements', *Quarterly Journal of the Royal Meteorological Society* **130**(598), 763–778.

URL: <https://doi.org/10.1256/qj.03.151>

Baran, A. J. (2009), 'A review of the light scattering properties of cirrus', *Journal of Quantitative Spectroscopy and Radiative Transfer* **110**(14-16), 1239–1260.

URL: <https://doi.org/10.1016/j.jqsrt.2009.02.026>

Baran, A. J., Hill, P., Furtado, K., Field, P. & Manners, J. (2014), 'A coupled cloud physics–radiation parameterization of the bulk optical properties of cirrus and its impact on the Met Office Unified Model global atmosphere 5.0 configuration', *Journal of Climate* **27**(20), 7725–7752.

URL: <http://doi.org/10.1175/JCLI-D-13-00700.1>

Baran, A. J., Watts, P. D. & Foot, J. S. (1998), 'Potential retrieval of dominating crystal habit and size using radiance data from a dual-view and multiwavelength instrument: A tropical cirrus anvil case', *Journal of Geophysical Research: Atmospheres* **103**(D6), 6075–6082.

URL: <http://doi.org/10.1029/97JD03122>

Baran, A. J., Yang, P. & Havemann, S. (2001), 'Calculation of the single-scattering properties of randomly oriented hexagonal ice columns: a comparison of the T-matrix and the finite-difference time-domain methods', *Applied Optics* **40**(24), 4376–4386.

URL: <https://doi.org/10.1364/AO.40.004376>

Baum, B. A., Yang, P., Heymsfield, A. J., Bansemer, A., Cole, B. H., Merrelli, A., Schmitt, C. & Wang, C. (2014), 'Ice cloud single-scattering property models with the full phase matrix at wavelengths from 0.2 to 100 μm ', *Journal of Quantitative Spectroscopy and Radiative Transfer* **146**, 123–139.

URL: <http://doi.org/10.1016/j.jqsrt.2014.02.029>

Baum, B. A., Yang, P., Nasiri, S., Heidinger, A. K., Heymsfield, A. & Li, J. (2007), 'Bulk scattering properties for the remote sensing of ice clouds. Part III: High-resolution spectral models from 100 to 3250 cm^{-1} ', *Journal of Applied Meteorology and Climatology* **46**(4), 423–434.

URL: <https://doi.org/10.1175/JAM2473.1>

- Beeby, R. (2011), Validation of the far-infrared foreign-broadened water vapour continuum from airborne field campaign measurements, PhD thesis, Imperial College London.
- Bellisario, C., Brindley, H. E., Murray, J. E., Last, A., Pickering, J., Harlow, R. C., Fox, S., Fox, C., Newman, S. M., Smith, M., Anderson, D., Huang, X. & Chen, X. (2017), 'Retrievals of the far infrared surface emissivity over the greenland plateau using the tropospheric airborne Fourier transform spectrometer (TAFTS)', *Journal of Geophysical Research: Atmospheres* **122**(22).
- URL: <http://dx.doi.org/10.1002/2017JD027328>
- Belotti, C., Barbara, F., Barucci, M., Bianchini, G., D'Amato, F., Del Bianco, S., Di Natale, G., Gai, M., Montori, A., Pratesi, F., Rettinger, M., Rolf, C., Sussmann, R., Trickl, T., Viciani, S., Vogelmann, H. & Palchetti, L. (2023), 'The Far-Infrared Radiation Mobile Observation System (FIRMOS) for spectral characterization of the atmospheric emission', *Atmospheric Measurement Techniques* **16**(10), 2511–2529.
- URL: <http://doi.org/10.5194/amt-16-2511-2023>
- Ben-Yami, M., Oetjen, H., Brindley, H., Cossich, W., Lajas, D., Maestri, T., Magurno, D., Raspollini, P., Sgheri, L. & Warwick, L. (2022), 'Emissivity retrievals with FORUM's end-to-end simulator: challenges and recommendations', *Atmospheric Measurement Techniques* **15**(6), 1755–1777.
- URL: <http://doi.org/10.5194/amt-15-1755-2022>
- Bergamaschi, P., Houweling, S., Segers, A., Krol, M., Frankenberg, C., Scheepmaker, R. A., Dlugokencky, E., Wofsy, S. C., Kort, E. A., Sweeney, C., Schuck, T., Brenninkmeijer, C., Chen, H., Beck, V. & Gerbig, C. (2013), 'Atmospheric CH₄ in the first decade of the 21st century: Inverse modeling analysis using sciamachy satellite retrievals and noaa surface measurements', *Journal of Geophysical Research: Atmospheres* **118**(13), 7350–7369.
- URL: <http://dx.doi.org/10.1002/jgrd.50480>
- Bianchini, G., Castagnoli, F., Di Natale, G. & Palchetti, L. (2019), 'A Fourier transform spectroradiometer for ground-based remote sensing of the atmospheric downwelling long-

wave radiance', *Atmospheric Measurement Techniques* **12**(1), 619–635.

URL: <https://doi.org/10.5194/amt-12-619-2019>

Brindley, H. E. & Harries, J. E. (1998), 'The impact of far i.r. absorption on clear sky greenhouse forcing: sensitivity studies at high spectral resolution', *Journal of Quantitative Spectroscopy and Radiative Transfer* **60**(2), 151–180.

URL: [https://doi.org/10.1016/S0022-4073\(97\)00152-0](https://doi.org/10.1016/S0022-4073(97)00152-0)

Brindley, H. E., Murray, J. E., Panditharatne, S., Warwick, L., Mosselmans, S., Belotti, C. & Huang, Y. (2025), 'W-band, HiSRAMS, AERI, FIRR2, FINESSE and FIRMOS Experiment on Remote Sensing (WHAFFERS) Campaign'. 'Last access: 30 May 2025'.

URL: <https://whafffers.meteo.mcgill.ca/>

Canas, T. A., Murray, J. E. & Harries, J. E. (1997), 'Tropospheric Airborne Fourier transform spectrometer (TAFTS)', *Satellite Remote Sensing of Clouds and the Atmosphere II* **3220**, 91 – 102.

URL: <https://doi.org/10.1117/12.301139>

Chahine, M. T., Pagano, T. S., Aumann, H. H., Atlas, R., Barnett, C., Blaisdell, J., Chen, L., Divakarla, M., Fetzer, E. J., Goldberg, M., Gautier, C., Granger, S., Hannon, S., Irion, F. W., Kakar, R., Kalnay, E., Lambrigtsen, B. H., Lee, S., Le Marshall, J., Mcmilan, W. W., Mcmillin, L., Olsen, E. T., Revercomb, H., Rosenkranz, P., Smith, W. L., Staelin, D., Strow, L. L., Susskind, J., Tobin, D., Wolf, W. & Zhou, L. (2006), 'AIRS: Improving weather forecasting and providing new data on greenhouse gases', *Bulletin of the American Meteorological Society* **87**(7), 911–926.

URL: <http://doi.org/10.1175/BAMS-87-7-911>

Chander, G., Hewison, T. J., Fox, N., Wu, X., Xiong, X. & Blackwell, W. J. (2013), 'Overview of intercalibration of satellite instruments', *IEEE Transactions on Geoscience and Remote Sensing* **51**(3), 1056–1080.

URL: <http://doi.org/10.1109/TGRS.2012.2228654>

Chepfer, H., Minnis, P., Young, D., Nguyen, L. & Arduini, R. F. (2002), 'Estimation of cirrus cloud effective ice crystal shapes using visible reflectances from dual-satellite measure-

ments', *Journal of Geophysical Research: Atmospheres* **107**(D23).

URL: <http://doi.org/10.1029/2000JD000240>

Chevallier, F., Michele, S. D. & McNally, A. (2006), Diverse profile datasets from the ECMWF 91-level short-range forecasts, Technical Report December, ECMWF.

URL: <https://www.ecmwf.int/node/8685>

Chou, M.-D., Lee, K.-T., Tsay, S.-C. & Fu, Q. (1999), 'Parameterization for cloud longwave scattering for use in atmospheric models', *Journal of Climate* **12**(1), 159 – 169.

URL: [https://doi.org/10.1175/1520-0442\(1999\)012%3C0159:PFCLSF%3E2.0.CO;2](https://doi.org/10.1175/1520-0442(1999)012%3C0159:PFCLSF%3E2.0.CO;2)

Chung, E.-S., Soden, B., Sohn, B. J. & Shi, L. (2014), 'Upper-tropospheric moistening in response to anthropogenic warming', *Proceedings of the National Academy of Sciences* **111**(32), 11636–11641.

URL: <http://doi.org/10.1073/pnas.1409659111>

Clough, S., Shephard, M., Mlawer, E., Delamere, J., Iacono, M., Cady-Pereira, K., Boukabara, S. & Brown, P. (2005), 'Atmospheric radiative transfer modeling: a summary of the AER codes', *Journal of Quantitative Spectroscopy and Radiative Transfer* **91**(2), 233–244.

URL: <https://doi.org/10.1016/j.jqsrt.2004.05.058>

Cohen, J., Zhang, X., Francis, J., Jung, T., Kwok, R., Overland, J., Ballinger, T. J., Bhatt, U. S., Chen, H. W., Coumou, D., Feldstein, S., Gu, H., Handorf, D., Henderson, G., Ionita, M., Kretschmer, M., Laliberte, F., Lee, S., Linderholm, H. W., Maslowski, W., Peings, Y., Pfeiffer, K., Rigor, I., Semmler, T., Stroeve, J., Taylor, P. C., Vavrus, S., Vihma, T., Wang, S., Wendisch, M., Wu, Y. & Yoon, J. (2019), 'Divergent consensus on arctic amplification influence on midlatitude severe winter weather', *Nature Climate Change* **10**(1), 20–29.

URL: <http://dx.doi.org/10.1038/s41558-019-0662-y>

Collard, A. D. (2007), Selection of IASI Channels for Use in Numerical Weather Prediction Selection of IASI Channels for Use in Numerical Weather Prediction, Technical report,

ECMWF.

URL: <https://doi.org/10.21957/60fy4u6fi>

Colman, R. & Soden, B. J. (2021), 'Water vapor and lapse rate feedbacks in the climate system', *Reviews of Modern Physics* **93**, 045002.

URL: <https://doi.org/10.1103/RevModPhys.93.045002>

Cox, C. V., Harries, J. E., Taylor, J. P., Green, P. D., Baran, A. J., Pickering, J. C., Last, A. E. & Murray, J. E. (2010), 'Measurement and simulation of mid- and far-infrared spectra in the presence of cirrus', *Quarterly Journal of the Royal Meteorological Society* **136**, 718–739.

URL: <https://doi.org/10.1002/QJ.596>

Cox, C. V., Murray, J. E., Taylor, J. P., Green, P. D., Pickering, J. C., Harries, J. E. & Last, A. E. (2007), 'Clear-sky far-infrared measurements observed with TAFTS during the EAQUATE campaign, September 2004', *Quarterly Journal of the Royal Meteorological Society* **133**(S3), 273–283.

URL: <https://doi.org/10.1002/QJ.159>

Cuesta-Valero, F. J., García-García, A., Beltrami, H., González-Rouco, J. F. & García-Bustamante, E. (2021), 'Long-term global ground heat flux and continental heat storage from geothermal data', *Climate of the Past* **17**(1), 451–468.

URL: <http://dx.doi.org/10.5194/cp-17-451-2021>

Davis, S. P., Abrams, M. C. & Brault, J. W. (2001), *Fourier Transform Spectrometry*, Academic Press.

Dessler, A. E., Zhang, Z. & Yang, P. (2008), 'Water-vapor climate feedback inferred from climate fluctuations, 2003–2008', *Geophysical Research Letters* **35**(L20704).

URL: <https://doi.org/10.1029/2008GL035333>

Di Natale, G., Bianchini, G., Del Guasta, M., Ridolfi, M., Maestri, T., Cossich, W., Magurno, D. & Palchetti, L. (2020), 'Characterization of the far infrared properties and radiative forcing of antarctic ice and water clouds exploiting the spectrometer-lidar synergy', *Remote Sensing*

12(21), 3574.

URL: <http://doi.org/10.3390/rs12213574>

Di Natale, G. & Palchetti, L. (2022), ‘Sensitivity studies toward the retrieval of ice crystal habit distributions inside cirrus clouds from upwelling far infrared spectral radiance observations’, *Journal of Quantitative Spectroscopy and Radiative Transfer* **282**, 108120.

URL: <https://doi.org/10.1016/j.jqsrt.2022.108120>

Di Natale, G., Palchetti, L., Bianchini, G. & Guasta, M. D. (2017), ‘Simultaneous retrieval of water vapour, temperature and cirrus clouds properties from measurements of far infrared spectral radiance over the antarctic plateau’, *Atmospheric Measurement Techniques* **10**(3), 825–837.

URL: <https://doi.org/10.5194/amt-10-825-2017>

Di Natale, G., Ridolfi, M. & Palchetti, L. (2024), ‘A new approach to crystal habit retrieval from far-infrared spectral radiance measurements’, *Atmospheric Measurement Techniques* **17**(10), 3171–3186.

URL: <http://doi.org/10.5194/amt-17-3171-2024>

Dils, B., Zhou, M., Camy-Peyret, C., De Mazière, M., Kangah, Y., Langerock, B., Prunet, P., Serio, C., Siddans, R. & Kerridge, B. (2024), ‘Independent validation of IASI/MetOp-A LMD and RAL CH₄ products using CAMS model, in situ profiles, and ground-based FTIR measurements’, *Atmospheric Measurement Techniques* **17**(18), 5491–5524.

URL: <http://doi.org/10.5194/amt-17-5491-2024>

DiNatale, G. & Palchetti, L. (2022), ‘Sensitivity studies toward the retrieval of ice crystal habit distributions inside cirrus clouds from upwelling far infrared spectral radiance observations’, *Journal of Quantitative Spectroscopy and Radiative Transfer* **282**, 108120.

URL: <https://doi.org/10.1016/j.jqsrt.2022.108120>

DiNatale, G., Palchetti, L., Bianchini, G. & Ridolfi, M. (2020), ‘The two-stream δ -Eddington approximation to simulate the far infrared earth spectrum for the simultaneous atmospheric

- and cloud retrieval', *Journal of Quantitative Spectroscopy and Radiative Transfer* **246**, 106927.
URL: <https://doi.org/10.1016/j.jqsrt.2020.106927>
- Dirksen, R. J., Sommer, M., Immler, F. J., Hurst, D. F., Kivi, R. & Vömel, H. (2014), 'Reference quality upper-air measurements: GRUAN data processing for the Vaisala RS92 radiosonde', *Atmospheric Measurement Techniques* **7**(12), 4463–4490.
URL: <https://doi.org/10.5194/amt-7-4463-2014>
- Dlugokencky, E., Crotwell, A., Lang, P., Higgs, J., Vaughn, B., Englund, S., Novelli, P., Wolter, S., Mund, J., Moglia, E. & Crotwell, M. (2021), Carbon cycle and greenhouse gases group flask-air sample measurements of CO₂, CH₄, CO, N₂O, H₂, SF₆ and isotopic ratios at global and regional background sites, 1967–present, version 2.0, NOAA national centers for environmental information, Technical report, NOAA ESRL.
- Dlugokencky, E. J., Mund, J. W., Crotwell, A. M., Crotwell, M. J. & Thoning, K. W. (2019), 'Atmospheric carbon dioxide dry air mole fractions from the NOAA GML carbon cycle cooperative global air sampling network, 1968-2019'. Version: 2019-07.
URL: <https://www.esrl.noaa.gov/gmd/ccgg/arc/?id=132>
- Ferreira, A. P., Nieto, R. & Gimeno, L. (2019), 'Completeness of radiosonde humidity observations based on the integrated global radiosonde archive', *Earth System Science Data* **11**(2), 603–627.
URL: <https://doi.org/10.5194/essd-11-603-2019>
- Fetzer, E. J., Read, W. G., Waliser, D., Kahn, B. H., Tian, B., Vömel, H., Irion, F. W., Su, H., Eldering, A., de la Torre Juarez, M., Jiang, J. & Dang, V. (2008), 'Comparison of upper tropospheric water vapor observations from the Microwave Limb Sounder and Atmospheric Infrared Sounder', *Journal of Geophysical Research: Atmospheres* **113**(D22110).
URL: <http://doi.org/10.1029/2008jd010000>
- Forster, P., Storelvmo, T., Armour, K., Collins, W., Dufresne, J.-L., Frame, D., Lunt, D., Mauritsen, T., Palmer, M., Watanabe, M., Wild, M. & Zhang, H. (2021), *Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the*

Intergovernmental Panel on Climate Change, Cambridge University Press. In Press.

URL: <https://doi.org/10.1017/9781009157896.009>

Fox, C., Green, P. D., Pickering, J. C. & Humpage, N. (2015), 'Analysis of far-infrared spectral radiance observations of the water vapor continuum in the arctic', *Journal of Quantitative Spectroscopy and Radiative Transfer* **155**, 57–65.

URL: <http://doi.org/10.1016/j.jqsrt.2015.01.001>

Fox, S., Mattioli, V., Turner, E., Vance, A., Cimini, D. & Gallucci, D. (2024), 'An evaluation of atmospheric absorption models at millimetre and sub-millimetre wavelengths using airborne observations', *Atmospheric Measurement Techniques* **17**(16), 4957–4978.

URL: <http://doi.org/10.5194/amt-17-4957-2024>

Fox, S., Mendrok, J., Eriksson, P., Ekelund, R., O'Shea, S. J., Bower, K. N., Baran, A. J., Harlow, R. C. & Pickering, J. C. (2019), 'Airborne validation of radiative transfer modelling of ice clouds at millimetre and sub-millimetre wavelengths', *Atmospheric Measurement Techniques* **12**(3), 1599–1617.

URL: <https://doi.org/10.5194/amt-12-1599-2019>

Fu, Y., Chen, Y., Li, R., Xian, T., Yu, L., Zhang, A., Liu, G. & Zhang, X. (2017), 'Lateral boundary of cirrus cloud from CALIPSO observations'.

URL: <https://doi.org/10.1038/s41598-017-14665-6>

Gelaro, R., McCarty, W., Suárez, M. J., Todling, R., Molod, A., Takacs, L., Randles, C. A., Darmenov, A., Bosilovich, M. G., Reichle, R., Wargan, K., Coy, L., Cullather, R., Draper, C., Akella, S., Buchard, V., Conaty, A., da Silva, A. M., Gu, W., Kim, G.-K., Koster, R., Lucchesi, R., Merkova, D., Nielsen, J. E., Partyka, G., Pawson, S., Putman, W., Rienecker, M., Schubert, S. D., Sienkiewicz, M. & Zhao, B. (2017), 'The Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2)', *Journal of Climate* **30**(14), 5419–5454.

URL: <https://doi.org/10.1175/jcli-d-16-0758.1>

Global Modeling and Assimilation Office & Pawson, S. (2015a), 'MERRA-2 inst1_2d_asm_nx:

2d,3-Hourly, Instantaneous, Single-Level, Assimilation, Single-Level diagnostics v5.12.4'.

URL: <https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/>

Global Modeling and Assimilation Office & Pawson, S. (2015b), 'MERRA-2 inst3_3d_asm_np: 3d,3-Hourly, Instantaneous, Pressure-Level, Assimilation, Assimilated Meteorological Fields V5.12.4'.

URL: <https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/>

Gordon, I., Rothman, L., Hargreaves, R., Hashemi, R., Karlovets, E., Skinner, F., Conway, E., Hill, C., Kochanov, R., Tan, Y., Wcisło, P., Finenko, A., Nelson, K., Bernath, P., Birk, M., Boudon, V., Campargue, A., Chance, K., Coustenis, A., Drouin, B., Flaud, J., Gamache, R., Hodges, J., Jacquemart, D., Mlawer, E., Nikitin, A., Perevalov, V., Rotger, M., Tennyson, J., Toon, G., Tran, H., Tyuterev, V., Adkins, E., Baker, A., Barbe, A., Canè, E., Császár, A., Dudaryonok, A., Egorov, O., Fleisher, A., Fleurbaey, H., Foltynowicz, A., Furtenbacher, T., Harrison, J., Hartmann, J., Horneman, V., Huang, X., Karman, T., Karns, J., Kass, S., Kleiner, I., Kofman, V., Kwabia-Tchana, F., Lavrentieva, N., Lee, T., Long, D., Lukashevskaya, A., Lyulin, O., Makhnev, V., Matt, W., Massie, S., Melosso, M., Mikhailenko, S., Mondelain, D., Müller, H., Naumenko, O., Perrin, A., Polyansky, O., Raddaoui, E., Raston, P., Reed, Z., Rey, M., Richard, C., Tóbiás, R., Sadiek, I., Schwenke, D., Starikova, E., Sung, K., Tamassia, F., Tashkun, S., Vander Auwera, J., Vasilenko, I., Vigasin, A., Villanueva, G., Vispoel, B., Wagner, G., Yachmenev, A. & Yurchenko, S. (2022), 'The HITRAN2020 molecular spectroscopic database', *Journal of Quantitative Spectroscopy and Radiative Transfer* **277**, 107949.

URL: <http://doi.org/10.1016/j.jqsrt.2021.107949>

Green, P. D., Newman, S. M., Beeby, R. J., Murray, J. E., Pickering, J. C. & Harries, J. E. (2012), 'Recent advances in measurement of the water vapour continuum in the far-infrared spectral region', *Trans. R. Soc. A* **370**, 2637–2655.

URL: <https://doi.org/10.1098/rsta.2011.0263>

Griffiths, P. R. & de Haseth, J. A. (1986), *Fourier Transform Infrared Spectrometry*, John Wiley & Sons.

Hakuba, M. Z., Fourest, S., Boyer, T., Meyssignac, B., Carton, J. A., Forget, G., Cheng, L.,

- Giglio, D., Johnson, G. C., Kato, S., Killick, R. E., Kolodziejczyk, N., Kuusela, M., Landerer, F., Llovel, W., Locarnini, R., Loeb, N., Lyman, J. M., Mishonov, A., Pilewskie, P., Reagan, J., Storto, A., Sukianto, T. & von Schuckmann, K. (2024), 'Trends and variability in earth's energy imbalance and ocean heat uptake since 2005', *Surveys in Geophysics* **45**(6), 1721–1756.
URL: <http://dx.doi.org/10.1007/s10712-024-09849-5>
- Hale, G. M. & Querry, M. R. (1973), 'Optical constants of water in the 200-nm to 200- μ m wavelength region', *Applied Optics* **12**(3), 555–563.
URL: <https://doi.org/10.1364/ao.12.000555>
- Hall, E. G., Jordan, A. F., Hurst, D. F., Oltmans, S. J., Vömel, H., Kühnreich, B. & Ebert, V. (2016), 'Advancements, measurement uncertainties, and recent comparisons of the NOAA frost point hygrometer', *Atmospheric Measurement Techniques* **9**(9), 4295–4310.
URL: <http://doi.org/10.5194/amt-9-4295-2016>
- Hanel, R. A., Schlachman, B., Clark, F. D., Prokesh, C. H., Taylor, J. B., Wilson, W. M. & Chaney, L. (1970), 'The Nimbus III Michelson interferometer', *Applied Optics* **9**(8), 1767.
URL: <http://doi.org/10.1364/AO.9.001767>
- Hanel, R. A., Schlachman, B., Rogers, D. & Vanous, D. (1971), 'Nimbus 4 Michelson interferometer', *Applied Optics* **10**(6), 1376.
URL: <http://doi.org/10.1364/AO.10.001376>
- Haywood, J., Donner, L., Jones, A. & Golaz, J.-C. (2009), *Global Indirect Radiative Forcing Caused by Aerosols: IPCC (2007) and Beyond*, The MIT Press, p. 451–468.
URL: <http://doi.org/10.7551/mitpress/8300.003.0021>
- Heymsfield, A. (1975), 'Cirrus uncinus generating cells and the evolution of cirriform clouds. Part I :aircraft observations of the growth of the ice phase', *Journal of the Atmospheric Sciences* **32**(4), 799–808.
URL: [https://doi.org/10.1175/1520-0469\(1975\)032<0799:CUGCAT>2.0.CO;2](https://doi.org/10.1175/1520-0469(1975)032<0799:CUGCAT>2.0.CO;2)

Heymsfield, A. J., Krämer, M., Luebke, A., Brown, P., Cziczo, D. J., Franklin, C., Lawson, P., Lohmann, U., McFarquhar, G., Ulanowski, Z. & Van Tricht, K. (2017), 'Cirrus Clouds', *Meteorological Monographs* **58**, 2.1–2.26.

URL: <https://doi.org/10.1175/amsmonographs-d-16-0010.1>

Hilton, F., Armante, R., August, T., Barnet, C., Bouchard, A., Camy-Peyret, C., Capelle, V., Clarisse, L., Clerbaux, C., Coheur, P.-F., Collard, A., Crevoisier, C., Dufour, G., Edwards, D., Fajjan, F., Fourrié, N., Gambacorta, A., Goldberg, M., Guidard, V., Hurtmans, D., Illingworth, S., Jacquinet-Husson, N., Kerzenmacher, T., Klaes, D., Lavanant, L., Masiello, G., Matricardi, M., McNally, A., Newman, S., Pavelin, E., Payan, S., Péquignot, E., Peyridieu, S., Phulpin, T., Remedios, J., Schlüssel, P., Serio, C., Strow, L., Stubenrauch, C., Taylor, J., Tobin, D., Wolf, W. & Zhou, D. (2012), 'Hyperspectral Earth observation from IASI: Five years of accomplishments', *Bulletin of the American Meteorological Society* **93**(3), 347–370.

URL: <http://doi.org/10.1175/BAMS-D-11-00027.1>

Hong, Y., Liu, G. & Li, J. L. (2016), 'Assessing the radiative effects of global ice clouds based on CloudSat and CALIPSO measurements', *Journal of Climate* **29**(21), 7651–7674.

URL: <https://doi.org/10.1175/JCLI-D-15-0799.1>

Huang, L., Mo, Z., Liu, L., Zeng, Z., Chen, J., Xiong, S. & He, H. (2021), 'Evaluation of hourly pwv products derived from ERA5 and MERRA-2 over the Tibetan Plateau using ground-based GNSS observations by two enhanced models', *Earth and Space Science* **8**.

URL: <https://doi.org/10.1029/2020EA001516>

Huang, X., Chen, X., Zhou, D. K. & Liu, X. (2016), 'An observationally based global band-by-band surface emissivity dataset for climate and weather simulations', *Journal of the Atmospheric Sciences* **73**, 3541–3555.

URL: <https://doi.org/10.1175/JAS-D-15-0355.1>

IPCC (2023), *The Earth's Energy Budget, Climate Feedbacks and Climate Sensitivity*, Cambridge University Press, p. 923–1054.

Jackson, R. C., McFarquhar, G. M., Fridlind, A. M. & Atlas, R. (2015), 'The dependence of

cirrus gamma size distributions expressed as volumes in $n_0 - \lambda - \mu$ phase space and bulk cloud properties on environmental conditions: Results from the small ice particles in cirrus experiment (sparticus)', *Journal of Geophysical Research: Atmospheres* **120**(19).

URL: <http://dx.doi.org/10.1002/2015JD023492>

Jiang, J. H., Su, H., Zhai, C., Wu, L., Minschwaner, K., Molod, A. M. & Tompkins, A. M. (2015), 'An assessment of upper troposphere and lower stratosphere water vapor in MERRA, MERRA2, and ECMWF reanalyses using Aura MLS observations', *Journal of Geophysical Research* **120**, 11,468–11,485.

URL: <https://doi.org/10.1002/2015JD023752>

Johnston, B. R., Randel, W. J. & Sjoberg, J. P. (2021), 'Evaluation of tropospheric moisture characteristics among COSMIC-2, ERA5, and MERRA-2 in the tropics and subtropics', *Remote Sensing* **13**, 1–20.

URL: <https://doi.org/10.3390/rs13050880>

Kärcher, B., Dörnbrack, A. & Sölch, I. (2014), 'Supersaturation variability and cirrus ice crystal size distributions', *Journal of the Atmospheric Sciences* **71**(8), 2905–2926.

URL: <https://doi.org/10.1175/JAS-D-13-0404.1>

Kempe, V., Oertel, D., Schuster, R., Becker-Ross, H. & Jahn, H. (1980), 'Absolute ir-spectra from the measurement of Fourier-spectrometers aboard Meteor 25 and 28', *Acta Astronautica* **7**(12), 1403–1416.

URL: [http://doi.org/10.1016/0094-5765\(80\)90015-6](http://doi.org/10.1016/0094-5765(80)90015-6)

Kratz, D. P., Mlynczak, M. G., Mertens, C. J., Brindley, H., Gordley, L. L., Martin-Torres, J., Miskolczi, F. M. & Turner, D. D. (2005), 'An inter-comparison of far-infrared line-by-line radiative transfer models', *Journal of Quantitative Spectroscopy and Radiative Transfer* **90**(3–4), 323–341.

URL: <http://doi.org/10.1016/j.jqsrt.2004.04.006>

Kuebbeler, M., Lohmann, U., Hendricks, J. & Kärcher, B. (2014), 'Dust ice nuclei effects on

cirrus clouds', *Atmospheric Chemistry and Physics* **14**(6), 3027–3046.

URL: <https://doi.org/10.5194/acp-14-3027-2014>

Kursinski, E. R. & Gebhardt, T. (2014), 'A method to deconvolve errors in GPS RO-derived water vapor histograms', *Journal of Atmospheric and Oceanic Technology* **31**(12), 2606–2628.

URL: <http://doi.org/10.1175/jtech-d-13-00233.1>

Lawson, P., O'Connor, D., Zmarzly, P., Weaver, K., Baker, B., Mo, Q. & Jonsson, H. (2006), 'The 2D-S (stereo) probe: Design and preliminary tests of a new airborne, high-speed, high-resolution particle imaging probe', *Journal of Atmospheric and Oceanic Technology* **23**(11), 1462–1477.

URL: <http://doi.org/10.1175/JTECH1927.1>

Libois, Q. & Blanchet, J. P. (2017), 'Added value of far-infrared radiometry for remote sensing of ice clouds', *Journal of Geophysical Research* **122**(12), 6541–6564.

URL: <https://doi.org/10.1002/2016JD026423>

Liou, K. N. (2002), *An Introduction to Atmospheric Radiation*, International Geophysics Series, 2 edn, Academic Press, San Diego, CA.

Loeb, N. G., Ham, S.-H., Allan, R. P., Thorsen, T. J., Meyssignac, B., Kato, S., Johnson, G. C. & Lyman, J. M. (2024), 'Observational assessment of changes in Earth's energy imbalance since 2000', *Surveys in Geophysics* **45**(6), 1757–1783.

URL: <http://dx.doi.org/10.1007/s10712-024-09838-8>

Lohmann, U., Lüönd, F. & Mahrt, F. (2016), *An Introduction to Clouds: From the Microscale to Climate*, Cambridge University Press.

URL: <http://doi.org/10.1017/CBO9781139087513>

Lohmann, U., Spichtinger, P., Jess, S., Peter, T. & Smit, H. (2008), 'Cirrus cloud formation and ice supersaturated regions in a global climate model', *Environmental Research Letters* **3**(4), 045022.

URL: <https://doi.org/10.1088/1748-9326/3/4/045022>

Lynch, D., Sassen, K., Starr, D. & Stephens, G. (2002), *Cirrus*, Oxford University Press.

L'Ecuyer, T. S., Drouin, B. J., Anheuser, J., Grames, M., Henderson, D. S., Huang, X., Kahn, B. H., Kay, J. E., Lim, B. H., Mateling, M., Merrelli, A., Miller, N. B., Padmanabhan, S., Peterson, C., Schlegel, N.-J., White, M. L. & Xie, Y. (2021), 'The Polar Radiant Energy in the Far Infrared Experiment: A new perspective on polar longwave energy exchanges', *Bulletin of the American Meteorological Society* **102**(7), E1431 – E1449.

URL: <https://doi.org/10.1175/BAMS-D-20-0155.1>

Maestri, T., Arosio, C., Rizzi, R., Palchetti, L., Bianchini, G. & Guasta, M. D. (2019), 'Antarctic ice cloud identification and properties using downwelling spectral radiance from 100 to 1,400 cm⁻¹', *Journal of Geophysical Research: Atmospheres* **124**(8), 4761–4781.

URL: <https://doi.org/10.1029/2018jd029205>

Maestri, T. & Holz, R. (2009), 'Retrieval of cloud optical properties from multiple infrared hyperspectral measurements: A methodology based on a line-by-line multiple-scattering code', *IEEE Transactions on Geoscience and Remote Sensing* **47**(8), 2413–2426.

URL: <http://doi.org/10.1109/TGRS.2009.2016105>

Maestri, T., Rizzi, R., Tosi, E., Veglio, P., Palchetti, L., Bianchini, G., Di Girolamo, P., Masiello, G., Serio, C. & Summa, D. (2014), 'Analysis of cirrus cloud spectral signatures in the far infrared', *Journal of Quantitative Spectroscopy and Radiative Transfer* **141**, 49–64.

URL: <http://doi.org/10.1016/j.jqsrt.2014.02.030>

Magurno, D., Cossich, W., Maestri, T., Bantges, R., Brindley, H., Fox, S., Harlow, C., Murray, J., Pickering, J., Warwick, L. & Oetjen, H. (2020), 'Cirrus cloud identification from airborne far-infrared and mid-infrared spectra', *Remote Sensing* **12**(13), 2097.

URL: <https://doi.org/10.3390/rs12132097>

Marenco, F. (2010), Capabilities and operation of the FAAM EZlidar, Technical report, Met Office OBR Tech.

Marenco, F., Johnson, B., Turnbull, K., Newman, S., Haywood, J., Webster, H. & Ricketts, H. (2011), 'Airborne lidar observations of the 2010 Eyjafjallajökull volcanic ash plume', *Journal*

of Geophysical Research: Atmospheres **116**(D20).

URL: <https://doi.org/10.1029/2011JD016396>

Marquardt, D. W. (1963), 'An algorithm for least-squares estimation of nonlinear parameters', *Journal of the Society for Industrial and Applied Mathematics* **11**(2), 431–441.

URL: <https://doi.org/10.1137/0111030>

Martinazzo, M., Magurno, D., Cossich, W., Serio, C., Masiello, G. & Maestri, T. (2021), 'Assessment of the accuracy of scaling methods for radiance simulations at far and mid infrared wavelengths', *Journal of Quantitative Spectroscopy and Radiative Transfer* **271**, 107739.

URL: <http://doi.org/10.1016/j.jqsrt.2021.107739>

Mast, J. C., Mlynczak, M. G., Cageao, R. P., Kratz, D. P., Latvakoski, H., Johnson, D. G., Turner, D. D. & Mlawer, E. J. (2017), 'Measurements of downwelling far-infrared radiance during the RHUBC-II campaign at Cerro Toco, Chile and comparisons with line-by-line radiative transfer calculations', *Journal of Quantitative Spectroscopy and Radiative Transfer* **198**, 25–39.

URL: <http://doi.org/10.1016/j.jqsrt.2017.04.028>

Masuda, K. (2006), 'Infrared sea surface emissivity including multiple reflection effect for isotropic gaussian slope distribution model', *Remote Sensing of Environment* **103**(4), 488–496.

URL: <https://doi.org/10.1016/j.rse.2006.04.011>

Masuda, K., Takashima, T. & Takayama, Y. (1988), 'Emissivity of pure and sea waters for the model sea surface in the infrared window regions', *Remote Sensing of Environment* **24**(2), 313–329.

URL: [http://doi.org/10.1016/0034-4257\(88\)90032-6](http://doi.org/10.1016/0034-4257(88)90032-6)

Matricardi, M. (2005), The inclusion of aerosols and clouds in RTIASI, the ECMWF fast radiative transfer model for the infrared atmospheric sounding interferometer., Technical report, ECMWF.

URL: <http://doi.org/10.21957/1krvb28ql>

Merrelli, A. & Turner, D. D. (2012), 'Comparing information content of upwelling far-infrared and mid infrared radiance spectra for clear atmosphere profiling', *Journal of Atmospheric*

and *Oceanic Technology* **29**(4), 510–526.

URL: <https://doi.org/10.1175/JTECH-D-11-00113.1>

Meyssignac, B., Ablain, M., Guérou, A., Prandi, P., Barnoud, A., Blazquez, A., Fourest, S., Rousseau, V., Bonnefond, P., Cazenave, A., Chenal, J., Dibarboue, G., Donlon, C., Benveniste, J., Sylvestre-Baron, A. & Vinogradova, N. (2023), ‘How accurate is accurate enough for measuring sea-level rise and variability’, *Nature Climate Change* **13**(8), 796–803.

URL: <http://dx.doi.org/10.1038/s41558-023-01735-z>

Miloshevich, L. M., Vömel, H., Whiteman, D. N. & Leblanc, T. (2009), ‘Accuracy assessment and correction of Vaisala RS92 radiosonde water vapor measurements’, *Journal of Geophysical Research* **114**(D11).

URL: <https://doi.org/10.1029/2008jd011565>

Mishchenko, M. I., Travis, L. D. & Mackowski, D. W. (1996), ‘T-matrix computations of light scattering by nonspherical particles: A review’, *Journal of Quantitative Spectroscopy and Radiative Transfer* **55**, 535–575.

Mlawer, E., Cady-Pereira, K., Mascio, J. & Gordon, I. (2023), ‘The inclusion of the MT_CKD water vapor continuum model in the HITRAN molecular spectroscopic database’, *Journal of Quantitative Spectroscopy and Radiative Transfer* **306**, 108645.

URL: <https://doi.org/10.1016/j.jqsrt.2023.108645>

Mlawer, E. J., Turner, D. D., Paine, S. N., Palchetti, L., Bianchini, G., Payne, V. H., Cady-Pereira, K. E., Pernak, R. L., Alvarado, M. J., Gombos, D., Delamere, J. S., Mlynczak, M. G. & Mast, J. C. (2019), ‘Analysis of water vapor absorption in the far-infrared and submillimeter regions using surface radiometric measurements from extremely dry locations’, *Journal of Geophysical Research: Atmospheres* **124**(14), 8134–8160.

URL: <https://doi.org/10.1029/2018JD029508>

Mlynczak, M. G., Johnson, D. G., Latvakoski, H., Jucks, K., Watson, M., Kratz, D. P., Bingham, G., Traub, W. A., Wellard, S. J., Hyde, C. R. & Liu, X. (2006), ‘First light from the far-infrared

spectroscopy of the troposphere (FIRST) instrument', *Geophysical Research Letters* **33**(7).

URL: <https://doi.org/10.1029/2005gl025114>

Mu, M., De Kauwe, M. G., Ukkola, A. M., Pitman, A. J., Guo, W., Hobeichi, S. & Briggs, P. R. (2021), 'Exploring how groundwater buffers the influence of heatwaves on vegetation function during multi-year droughts', *Earth System Dynamics* **12**(3), 919–938.

URL: <http://dx.doi.org/10.5194/esd-12-919-2021>

Murray, J. (2019), TAFTS: Overview of performance during PIKNMIX-F [European Space Agency Report Reference Number: FORME-ICL-TN1], Technical report, Space and Atmospheric Physics Group, Imperial College London.

Murray, J. E., Brindley, H. E., Fox, S., Bellisario, C., Pickering, J. C., Fox, C., Harlow, C., Smith, M., Anderson, D., Huang, X., Chen, X., Last, A. & Bantges, R. (2020), 'Retrievals of high-latitude surface emissivity across the infrared from high-altitude aircraft flights', *Journal of Geophysical Research: Atmospheres* **125**(22).

URL: <https://doi.org/10.1029/2020jd033672>

Murray, J. E., Warwick, L., Brindley, H., Last, A., Quigley, P., Rochester, A., Dewar, A. & Cummins, D. (2024), 'The Far-INfrarEd Spectrometer for Surface Emissivity (FINESSE) – Part 1: Instrument description and level 1 radiances', *Atmospheric Measurement Techniques* **17**(16), 4757–4775.

URL: <http://doi.org/10.5194/amt-17-4757-2024>

Naethe, P., Asgari, M., Kneer, C., Knieps, M., Jenal, A., Weber, I., Moelter, T., Dzunic, F., Deffert, P., Rommel, E., Delaney, M., Baschek, B., Rock, G., Bongartz, J. & Burkart, A. (2023), 'Calibration and validation from ground to airborne and satellite level: Joint application of time-synchronous field spectroscopy, drone, aircraft and Sentinel-2 imaging', *PFG – Journal of Photogrammetry, Remote Sensing and Geoinformation Science* **91**(1), 43–58.

URL: <http://doi.org/10.1007/s41064-022-00231-x>

Newman, S. M., Larar, A. M., Smith, W. L., Ptashnik, I. V., Jones, R. L., Mead, M. I., Revercomb, H., Tobin, D. C., Taylor, J. K. & Taylor, J. P. (2012), 'The joint airborne IASI validation experi-

ment: An evaluation of instrument and algorithms', *Journal of Quantitative Spectroscopy and Radiative Transfer* **113**(11), 1372–1390.

URL: <http://doi.org/10.1016/j.jqsrt.2012.02.030>

Niro, F., Goryl, P., Dransfeld, S., Boccia, V., Gascon, F., Adams, J., Themann, B., Scifoni, S. & Doxani, G. (2021), 'European Space Agency (ESA) calibration/validation strategy for optical land-imaging satellites and pathway towards interoperability', *Remote Sensing* **13**(15), 3003.

URL: <http://doi.org/10.3390/rs13153003>

Norton, R. H. & Beer, R. (1976), 'New apodizing functions for Fourier spectrometry', *J. Opt. Soc. Am.* **66**(3), 259–264.

URL: <https://doi.org/10.1364/JOSA.66.000259>

Nussenzveig, H. M. & Wiscombe, W. J. (1980), 'Efficiency factors in Mie scattering', *Phys. Rev. Lett.* **45**, 1490–1494.

URL: <https://doi.org/10.1103/PhysRevLett.45.1490>

Oetjen, H., Brindley, H., Buehler, S. A., Clarisse, L., Coppens, D., Deschamps, A., Dinelli, B. M., Harrison, J. J., Labonnote, L., Libois, Q., Mlynchak, M., Palchetti, L., Ridolfi, M., Riese, M. & Saunders, R. (2020), Far-infrared-Outgoing-Radiation Understanding and Monitoring (FORUM) - Mission requirements document, Technical report, European Space Agency.

Oreopoulos, L., Mlawer, E., Delamere, J., Shippert, T., Cole, J., Fomin, B., Iacono, M., Jin, Z., Li, J., Manners, J., Räisänen, P., Rose, F., Zhang, Y., Wilson, M. J. & Rossow, W. B. (2012), 'The continual intercomparison of radiation codes: Results from Phase I', *Journal of Geophysical Research: Atmospheres* **117**(D6).

URL: <http://doi.org/10.1029/2011JD016821>

O'Shea, S. J., Choularton, T. W., Lloyd, G., Crosier, J., Bower, K. N., Gallagher, M., Abel, S. J., Cotton, R. J., Brown, P. R. A., Fugal, J. P., Schlenczek, O., Borrmann, S. & Pickering, J. C. (2016), 'Airborne observations of the microphysical structure of two contrasting cirrus

clouds', *Journal of Geophysical Research.-Atmos* p. 13510–13536.

URL: <https://doi.org/10.1002/2016JD025278>

O'Shea, S., Crosier, J., Dorsey, J., Gallagher, L., Schledewitz, W., Bower, K., Schlenczek, O., Borrmann, S., Cotton, R., Westbrook, C. & Ulanowski, Z. (2021), 'Characterising optical array particle imaging probes: implications for small-ice-crystal observations', *Atmospheric Measurement Techniques* **14**(3), 1917–1939.

URL: <http://doi.org/10.5194/amt-14-1917-2021>

Palchetti, L., Barucci, M., Belotti, C., Bianchini, G., Cluzet, B., D'Amato, F., Bianco, S. D., Di Natale, G., Gai, M., Khordakova, D., Montori, A., Oetjen, H., Rettinger, M., Rolf, C., Schuettemeyer, D., Sussmann, R., Viciani, S., Vogelmann, H. & Wienhold, F. G. (2021), 'Observations of the downwelling far-infrared atmospheric emission at the Zugspitze observatory', *Earth System Science Data* **13**(9), 4303–4312.

URL: <https://doi.org/10.5194/essd-13-4303-2021>

Palchetti, L., Brindley, H., Bantges, R., Buehler, S. A., Camy-Peyret, C., Carli, B., Cortesi, U., Bianco, S. D., Natale, G. D., Dinelli, B. M., Feldman, D., Huang, X. L., C.-Labonnote, L., Libois, Q., Maestri, T., Mlynczak, M. G., Murray, J. E., Oetjen, H., Ridolfi, M., Riese, M., Russell, J., Saunders, R. & Serio, C. (2020), 'FORUM: unique far-infrared satellite observations to better understand how Earth radiates energy to space', *American Meteorological Society* **101**(12), 2030–2046.

URL: <https://doi.org/10.1175/BAMS-D-19-0322.1>

Panditharatne, S., Brindley, H., Cox, C., Siddans, R., Murray, J., Warwick, L. & Fox, S. (2025), 'Retrievals of water vapour and temperature exploiting the far-infrared: application to aircraft observations in preparation for the FORUM mission', *Atmospheric Measurement Techniques* **18**(3), 717–735.

URL: <http://doi.org/10.5194/amt-18-717-2025>

Panditharatne, S., Brindley, H., Cox, C., Song, R., Siddans, R., Murray, J., Fox, S. & Fox, C. (under review), 'Exploiting airborne far-infrared measurements to optimise an ice cloud retrieval', *Atmospheric Chemistry and Physics* .

- Peterson, C. A., Huang, X., Chen, X. & Yang, P. (2022), 'Synergistic use of far- and mid-infrared spectral radiances for satellite-based detection of polar ice clouds over ocean', *Journal of Geophysical Research: Atmospheres* **127**(9), e2021JD035733.
URL: <https://doi.org/10.1029/2021JD035733>
- Pfreundschuh, S., Fox, S., Eriksson, P., Duncan, D., Buehler, S. A., Brath, M., Cotton, R. & Ewald, F. (2022), 'Synergistic radar and sub-millimeter radiometer retrievals of ice hydrometeors in mid-latitude frontal cloud systems', *Atmospheric Measurement Techniques* **15**(3), 677–699.
URL: <http://doi.org/10.5194/amt-15-677-2022>
- Platnick, S., King, M. D., Ackerman, S. A., Menzel, W. P., Baum, B. A., Riédi, J. C. & Frey, R. A. (2003), 'The MODIS cloud products: Algorithms and examples from Terra', *IEEE Transactions on geoscience and Remote Sensing* **41**(2), 459–473.
URL: <https://doi.org/10.1109/TGRS.2002.808301>
- Rabier, F., Fourrié, N., Chafäi, D. & Prunet, P. (2002), 'Channel selection methods for infrared atmospheric sounding interferometer radiances', *Quarterly Journal of the Royal Meteorological Society* **128**(581), 1011–1027.
URL: <https://doi.org/10.1256/0035900021643638>
- Raghuraman, S. P., Paynter, D. & Ramaswamy, V. (2021), 'Anthropogenic forcing and response yield observed positive trend in Earth's energy imbalance', *Nature Communications* **12**(4577).
URL: <https://doi.org/10.1038/S41467-021-24544-4>
- Ridolfi, M., Bianco, S. D., Roma, A. D., Castelli, E., Belotti, C., Dandini, P., Di Natale, G., Dinelli, B. M., Labonnote, L. C. & Palchetti, L. (2020), 'FORUM Earth Explorer 9: Characteristics of level 2 products and synergies with IASI-NG', *Remote Sensing* **12**, 1496.
URL: <https://doi.org/10.3390/rs12091496>
- Rodgers, C. D. (2000), *Inverse Methods for Atmospheric Sounding*, Vol. 2, World Scientific Publishing.
URL: <http://doi.org/10.1142/3171>

- Saito, M., Iwabuchi, H., Yang, P., Tang, G., King, M. D. & Sekiguchi, M. (2017), 'Ice particle morphology and microphysical properties of cirrus clouds inferred from combined caliop-
iir measurements', *Journal of Geophysical Research: Atmospheres* **122**(8), 4440–4462.
URL: <http://doi.org/10.1002/2016JD026080>
- Salby, M. L. (1996), *Fundamentals of Atmospheric Physics*, Vol. 61 of *International Geophysics Series*, Academic Press Inc.
- Santer, B. D., Po-Chedley, S., Mears, C., Fyfe, J. C., Gillett, N., Fu, Q., Painter, J. F., Solomon, S., Steiner, A. K., Wentz, F. J., Zelinka, M. D. & Zou, C.-Z. (2021), 'Using climate model simulations to constrain observations', *Journal of Climate* **34**(15), 6281–6301.
URL: <http://doi.org/10.1175/JCLI-D-20-0768.1>
- Sassen, K., Wang, Z. & Liu, D. (2008), 'Global distribution of cirrus clouds from CloudSat/Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) measurements', *Journal of Geophysical Research: Atmospheres* **113**(D8).
URL: <http://doi.org/10.1029/2008JD009972>
- Saunders, R., Hocking, J., Rundle, D., Rayer, P., Havemann, S., Matricardi, M., Geer, A., Lupu, C., Brunel, P. & Vidot, J. (2017), RTTOV-12 Science and Validation Report, Technical report, EUMETSAT.
URL: https://nwp-saf.eumetsat.int/site/download/documentation/rtm/docs_rttov12/rttov12_svr.pdf
- Saunders, R., Hocking, J., Turner, E., Rayer, P., Rundle, D., Brunel, P., Vidot, J., Roquet, P., Matricardi, M., Geer, A., Bormann, N. & Lupu, C. (2018), 'An update on the rttov fast radiative transfer model (currently at version 12)', *Geoscientific Model Development* **11**(7), 2717–2737.
URL: <http://dx.doi.org/10.5194/gmd-11-2717-2018>
- Schröder, M., Lockhoff, M., Shi, L., August, T., Bennartz, R., Brogniez, H., Calbet, X., Fell, F., Forsythe, J., Gambacorta, A., Ho, S.-p., Kursinski, E. R., Reale, A., Trent, T. & Yang, Q. (2019), 'The GEWEX Water Vapor Assessment: Overview and introduction to results and

recommendations', *Remote Sensing* **11**(3).

URL: <https://doi.org/10.3390/rs11030251>

Seeman, S., Borbas, E. E., Knuteson, R., Stephenson, G. R. & Huang, H. (2008), 'Development of a global infrared land surface emissivity database for application to clear sky sounding retrievals from multispectral satellite radiance measurements', *Journal of Applied Meteorology and Climatology* **47**(1), 108–123.

URL: <https://doi.org/10.1175/2007jamc1590.1>

Serio, C., Masiello, G., Esposito, F., Di Girolamo, P., Di Iorio, T., Palchetti, L., Bianchini, G., Muscari, G., Pavese, G., Rizzi, R., Carli, B. & Cuomo, V. (2008), 'Retrieval of foreign-broadened water vapor continuum coefficients from emitted spectral radiance in the H₂O rotational band from 240 to 590 cm⁻¹', *Optics Express* **16**(20), 15816.

URL: <http://doi.org/10.1364/OE.16.015816>

Sherwood, S. C., Roca, R., Weckwerth, T. M. & Andronova, N. G. (2010), 'Tropospheric water vapor, convection, and climate', *Reviews of Geophysics* **48**(2).

URL: <http://dx.doi.org/10.1029/2009RG000301>

Shi, L., Schreck III, C. J., John, V. O., Chung, E.-S., Lang, T., Buehler, S. A. & Soden, B. J. (2022), 'Assessing the consistency of satellite-derived upper tropospheric humidity measurements', *Atmospheric Measurement Techniques* **15**(23), 6949–6963.

URL: <https://doi.org/10.5194/amt-15-6949-2022/>

Siddans, R. (2019), Water Vapour Climate Change Initiative (WV_cci) - Phase One, Deliverable 2.2; Version 1.0, RAL Space Remote Sensing Group-Technical Report, Technical report, RAL Space.

URL: https://climate.esa.int/documents/2497/Water_Vapour_CCI_D2.2_ATBD_Part2-IMS_L2_product_v2.0.pdf

Singh, D. K., Sanyal, S. & Wuebbles, D. J. (2024), 'Understanding the role of contrails and contrail cirrus in climate change: a global perspective', *Atmospheric Chemistry and Physics*

24(16), 9219–9262.

URL: <http://doi.org/10.5194/acp-24-9219-2024>

Sinha, A. & Harries, J. E. (1995), 'Water vapour and greenhouse trapping: The role of far infrared absorption', *Geophysical Research Letters* **22**(16), 2147–2150.

URL: <https://doi.org/10.1029/95GL01891>

Slater, T., Lawrence, I. R., Otosaka, I. N., Shepherd, A., Gourmelen, N., Jakob, L., Tepes, P., Gilbert, L. & Nienow, P. (2021), 'Review article: Earth's ice imbalance', *The Cryosphere* **15**(1), 233–246.

URL: <http://dx.doi.org/10.5194/tc-15-233-2021>

Stamnes, K., Tsay, S.-C. & Lazlo, I. (2000), DISORT, a general-purpose fortran program for Discrete-Ordinate-Method Radiative Transfer in scattering and emitting layered media: Documentation of methodology, Technical report, Stevens Institute of Technology.

URL: <https://www.libradtran.org/lib/exe/fetch.php?media=disortreport1.1.pdf>

Stubenrauch, C. J., Chédin, A., Rädcl, G., Scott, N. A. & Serrar, S. (2006), 'Cloud properties and their seasonal and diurnal variability from tovs path-b', *Journal of Climate* **19**(21), 5531–5553.

URL: <http://doi.org/10.1175/JCLI3929.1>

Sun, B., Calbet, X., Reale, A., Schroeder, S., Bali, M., Smith, R. & Pettey, M. (2021), 'Accuracy of Vaisala RS41 and RS92 upper tropospheric humidity compared to satellite hyperspectral infrared measurements', *Remote Sensing* **13**(2), 173.

URL: <http://doi.org/10.3390/rs13020173>

Trent, T., Schröder, M., Ho, S.-P., Beirle, S., Bennartz, R., Borbas, E., Borger, C., Brogniez, H., Calbet, X., Castelli, E., Compo, G. P., Ebisuzaki, W., Falk, U., Fell, F., Forsythe, J., Hersbach, H., Kachi, M., Kobayashi, S., Kursinski, R. E., Loyola, D., Luo, Z., Nielsen, J. K., Papandrea, E., Picon, L., Preusker, R., Reale, A., Shi, L., Slivinski, L., Teixeira, J., Vonder Haar, T. & Wagner, T. (2024), 'Evaluation of total column water vapour products from satellite observations and reanalyses within the GEWEX water vapor assessment', *Atmospheric*

Chemistry and Physics **24**(16), 9667–9695.

URL: <http://doi.org/10.5194/acp-24-9667-2024>

Trent, T., Schröder, M. & Remedios, J. (2019), ‘GEWEX Water Vapor Assessment: Validation of AIRS tropospheric humidity profiles with characterized radiosonde soundings’, *Journal of Geophysical Research: Atmospheres* **124**(2), 886–906.

URL: <http://doi.org/10.1029/2018JD028930>

Trent, T., Siddens, R., Kerridge, B., Schroeder, M., Scott, N. A. & Remedios, J. (2023), ‘Evaluation of tropospheric water vapour and temperature profiles retrieved from Metop-A by the Infrared and Microwave Sounding scheme’, *Atmospheric Measurement Techniques* .

URL: <https://doi.org/10.5194/amt-16-1503-2023>

Turetsky, M. R., Abbott, B. W., Jones, M. C., Anthony, K. W., Olefeldt, D., Schuur, E. A. G., Grosse, G., Kuhry, P., Hugelius, G., Koven, C., Lawrence, D. M., Gibson, C., Sannel, A. B. K. & McGuire, A. D. (2020), ‘Carbon release through abrupt permafrost thaw’, *Nature Geoscience* **13**(2), 138–143.

URL: <http://dx.doi.org/10.1038/s41561-019-0526-0>

Turner, D. & Holz, R. (2005), ‘Retrieving cloud fraction in the field-of-view of a high-spectral resolution infrared radiometer’, *IEEE Geoscience and Remote Sensing Letters* **2**(3), 287–291.

URL: <https://doi.org/10.1109/LGRS.2005.850533>

van Diedenhoven, B. & Kokhanovsky, A. (2018), *Remote sensing of crystal shapes in ice clouds*, Springer International.

URL: https://doi.org/10.1007/978-3-319-70808-9_5

Vömel, H., Naebert, T., Dirksen, R. & Sommer, M. (2016), ‘An update on the uncertainties of water vapor measurements using cryogenic frost point hygrometers’, *Atmospheric Measurement Techniques* **9**(8), 3755–3768.

URL: <http://doi.org/10.5194/amt-9-3755-2016>

von Schuckmann, K., Minière, A., Gues, F., Cuesta-Valero, F. J., Kirchengast, G., Adusumilli, S., Straneo, F., Ablain, M., Allan, R. P., Barker, P. M., Beltrami, H., Blazquez, A., Boyer, T.,

Cheng, L., Church, J., Desbruyeres, D., Dolman, H., Domingues, C. M., García-García, A., Giglio, D., Gilson, J. E., Gorfer, M., Haimberger, L., Hakuba, M. Z., Hendricks, S., Hosoda, S., Johnson, G. C., Killick, R., King, B., Kolodziejczyk, N., Korosov, A., Krinner, G., Kuusela, M., Landerer, F. W., Langer, M., Lavergne, T., Lawrence, I., Li, Y., Lyman, J., Marti, F., Marzeion, B., Mayer, M., MacDougall, A. H., McDougall, T., Monselesan, D. P., Nitzbon, J., Otosaka, I., Peng, J., Purkey, S., Roemmich, D., Sato, K., Sato, K., Savita, A., Schweiger, A., Shepherd, A., Seneviratne, S. I., Simons, L., Slater, D. A., Slater, T., Steiner, A. K., Suga, T., Szekely, T., Thiery, W., Timmermans, M.-L., Vanderkelen, I., Wjiffels, S. E., Wu, T. & Zemp, M. (2023), 'Heat stored in the earth system 1960–2020: where does the energy go?', *Earth System Science Data* **15**(4), 1675–1709.

URL: <http://dx.doi.org/10.5194/essd-15-1675-2023>

Wang, C., Yang, P., Baum, B. A., Platnick, S., Heidinger, A. K., Hu, Y. & Holz, R. E. (2011), 'Retrieval of ice cloud optical thickness and effective particle size using a fast infrared radiative transfer model', *J. Appl. Meteorol. Climatol.* **50**(11), 2283–2297.

Wang, S., Ren, T., Yang, P., Saito, M. & Brindley, H. E. (2024), 'Improved temperature-dependent ice refractive index compilation in the far-infrared spectrum', *Geophysical Research Letters* **51**(14).

URL: <http://doi.org/10.1029/2024GL110176>

Warren, S. G. & Brandt, R. E. (2008), 'Optical constants of ice from the ultraviolet to the microwave: A revised compilation', *Journal of Geophysical Research* **113**, 14220.

URL: <https://doi.org/10.1029/2007JD009744>

Warwick, L., Brindley, H., Roma, A. D., Fox, S., Havemann, S., Murray, J., Oetjen, H., Price, H. C., Schüttemeyer, D., Sgheri, L. & Tiddeman, D. A. (2022), 'Retrieval of tropospheric water vapor from airborne far-infrared measurements: A case study', *Journal of Geophysical Research: Atmospheres* **127**(7).

URL: <https://doi.org/10.1029/2020jd034229>

Warwick, L., Murray, J. E. & Brindley, H. (2024), 'The Far-INfrarEd Spectrometer for Surface Emissivity (FINESSE) – Part 2: First measurements of the emissivity of water in the far-

infrared', *Atmospheric Measurement Techniques* **17**(16), 4777–4787.

URL: <http://doi.org/10.5194/amt-17-4777-2024>

Wen, S. & Rose, W. I. (1994), 'Retrieval of sizes and total masses of particles in volcanic clouds using AVHRR bands 4 and 5', *J. Geophys. Res.* **99**(D3), 5421–5431.

Wendisch, M., Yang, P. & Pilewskie, P. (2007), 'Effects of ice crystal habit on thermal infrared radiative properties and forcing of cirrus', *Journal of Geophysical Research* **112**, 8201.

URL: <http://doi.org/10.1029/2006JD007899>

Wernberg, T., Thomsen, M. S., Baum, J. K., Bishop, M. J., Bruno, J. F., Coleman, M. A., Filbee-Dexter, K., Gagnon, K., He, Q., Murdiyarso, D., Rogers, K., Silliman, B. R., Smale, D. A., Starko, S. & Vanderklift, M. A. (2024), 'Impacts of climate change on marine foundation species', *Annual Review of Marine Science* **16**(1), 247–282.

URL: <http://dx.doi.org/10.1146/annurev-marine-042023-093037>

Weston, P. (2011), 'Progress towards the implementation of correlated observation errors in 4D-Var'. Met Office Forecasting Research Technical Report.

Wilson, S. H. S., Atkinson, N. C. & Smith, J. A. (1999), 'The development of an airborne infrared interferometer for meteorological sounding studies', *Journal of Atmospheric and Oceanic Technology* **16**(12), 1912–1927.

URL: [https://doi.org/10.1175/1520-0426\(1999\)016<1912:tdoaaai>2.0.co;2](https://doi.org/10.1175/1520-0426(1999)016<1912:tdoaaai>2.0.co;2)

Yang, P., Bi, L., Baum, B. A., Liou, K.-N., Kattawar, G. W., Mishchenko, M. I. & Cole, B. (2013), 'Spectrally consistent scattering, absorption, and polarization properties of atmospheric ice crystals at wavelengths from 0.2 to 100 μm ', *J. Atmos. Sci.* **70**(1), 330–347.

Yang, P., Liou, K.-N., Bi, L., Liu, C., Yi, B. & Baum, B. A. (2015), 'On the radiative properties of ice clouds: Light scattering, remote sensing, and radiation parameterization', *Advances in Atmospheric Sciences* **32**(1), 32–63.

URL: <http://doi.org/10.1007/s00376-014-0011-z>

- Yang, P., Wei, H., Huang, H.-L., Baum, B. A., Hu, Y. X., Kattawar, G. W., Mishchenko, M. I. & Fu, Q. (2005), 'Scattering and absorption property database for nonspherical ice particles in the near- through far-infrared spectral region', *Applied Optics* **44**, 5512–5523.
URL: <https://doi.org/10.1364/AO.44.005512>
- Zhang, Y., Macke, A. & Albers, F. (1999), 'Effect of crystal size spectrum and crystal shape on stratiform cirrus radiative forcing', *Atmospheric Research* **52**(1), 59–75.
URL: [https://doi.org/10.1016/S0169-8095\(99\)00026-5](https://doi.org/10.1016/S0169-8095(99)00026-5)
- Zhao, B., Wang, Y., Gu, Y., Liou, K.-N., Jiang, J. H., Fan, J., Liu, X., Huang, L. & Yung, Y. L. (2019), 'Ice nucleation by aerosols from anthropogenic pollution', *Nat. Geosci.* **12**(8), 602–607.
URL: <https://doi.org/10.1038/s41561-019-0389-4>
- Zhao, Y., Mace, G. G. & Comstock, J. M. (2011), 'The occurrence of particle size distribution bimodality in midlatitude cirrus as inferred from ground-based remote sensing data', *Journal of the Atmospheric Sciences* **68**(6), 1162–1177.
URL: <http://dx.doi.org/10.1175/2010JAS3354.1>
- Zhou, C., Dessler, A. E., Zelinka, M. D., Yang, P. & Wang, T. (2014), 'Cirrus feedback on interannual climate fluctuations', *Geophysical Research Letters* **41**(24), 9166–9173.
URL: <https://doi.org/10.1002/2014GL062095>