

Space and Atmospheric Physics Group
Department of Physics
Imperial College of Science, Technology and Medicine

Measuring tropospheric water vapour and surface emissivity using far-infrared radiances

Laura Rachel Warwick

Supervised by Prof. Helen Brindley and Dr. Paul Green

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Abstract

This thesis describes two strands of work relating to the gathering of far-infrared (wavenumbers $10\text{--}667\text{ cm}^{-1}$ wavelengths $15\text{--}1000\mu\text{m}$) radiance measurements, exploring the capabilities of the instruments involved and examining the data generated in the context of the relevant models.

The first strand of the thesis explores the role of atmospheric water vapour which absorbs strongly in the far-infrared. The work described makes use of spectrally resolved far-infrared radiance measurements taken during the PIKNMIX-F airborne field campaign. On this field campaign co-incident upwelling mid- and far-infrared spectra were recorded in clear sky and overflying cirrus. The clear sky spectra from one flight are used to investigate the sensitivity of the far-infrared to the atmospheric water profile. Forward modelling is used to explore the changes to the expected radiance caused by changes in the surface properties and water vapour spectroscopy. Retrievals of water vapour and temperature profiles are also carried out. These show that the far-infrared radiance measurements contain more information about the atmospheric water vapour profile than the co-incident mid-infrared radiance measurements for this set of instruments.

The second strand of the thesis describes the new FINESSE instrument and its first measurements of far-infrared surface emissivity. FINESSE combines a commercial Fourier transform spectrometer with a spectral range of $400\text{--}1600\text{ cm}^{-1}$ and a custom pointing and calibration system. The main purpose of FINESSE is to make in-situ measurements of surface emissivity extending into the far-infrared. As part of the development of FINESSE, a simulator is produced that allows for the emissivity retrieval to be tested under different environmental conditions. This thesis describes the first measurements of emissivity made by FINESSE. The emissivity of deionised water is retrieved using radiance measurements made from the rooftop of Imperial College London during summer and winter conditions. The measurements compare favourably to theoretical calculations and previous mid-infrared measurements.

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I dedicate this thesis to my family: Mum, Dad and Joe, and to Jake.

Statement of Originality

I confirm that the following thesis is my own work and that all information from the published and unpublished work of others has been acknowledged and referenced.

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Abbreviations

ABB ambient black body

AIRS Atmospheric Infrared Sounder

ARI Absolute Radiance Interferometer

ASTER Advanced Spaceborne Thermal Emission Reflection Radiometer

CESM Community Earth System Model

CLARREO Climate Absolute Radiance and Refractivity Observatory

DFS degrees of freedom for signal

ECOSTRESS ECOsystem and Spaceborne Thermal Radiometer Experiment on Space Station

ESA European Space Agency

EVI Earth Venture Instrument

FAAM Facility for Airborne Atmospheric Measurements

FINESSE Far INfrarEd Spectrometer for Surface Emissivity

FIR far-infrared

FIRMOS Far-Infrared Radiation Mobile Observation System

FORUM Far Infrared Outgoing Radiation Understanding and Monitoring

GRUAN Global Climate Observing System Reference Upper Air Network

HBB hot black body

HITRAN High-resolution Transmission Molecular Absorption

IASI Infrared Atmospheric Sounding Interferometer

ILS instrument line shape

IPCC Intergovernmental Panel on Climate Change

LBLRTM Line-By-Line Radiative Transfer Model

MCT Mercury-Cadmium-Telluride

MIR mid-infrared

MT_CKD Mlawer Tobin Clough Kneizys Davies (continuum model)

NASA National Aeronautics and Space Administration

NESR noise equivalent spectral radiance

OLR outgoing longwave radiation

OPD optical path difference

PREFIRE Polar Radiant Energy in the Far Infrared Experiment

PRT platinum resistance thermometer

REFIR—PAD Radiation Explorer in the Far InfraRed—Prototype of Applications and Development

RTTOV Radiative Transfer for TOVS

SEVIRI Spinning Enhanced Visible and InfraRed Imager

SLR straight and level run

TAFTS Tropospheric Airborne Fourier Transform Spectrometer

TICFIRE Thin Ice Clouds in the Far Infrared Experiment

TOA top of the atmosphere

UM Unified Model

Chapter 1

Introduction

This thesis brings together two strands of work exploring the utilisation of spectrally resolved far-infrared (FIR) radiance measurements for the understanding of Earth’s climate. The first strand focuses on understanding the atmospheric water vapour profile using measurements from an airborne spectrometer and the second strand describes the development and first data from a spectrometer designed to make measurements of surface emissivity in the mid-infrared (MIR) and FIR. The studies are particularly timely because of the recent selection of two satellite missions making spectrally resolved FIR radiance measurements. Of particular interest to this thesis is the Far Infrared Outgoing Radiation Understanding and Monitoring (FORUM) mission, the European Space Agency’s 9th Earth Explorer (ESA, 2019). FORUM will make comprehensive spectrally resolved measurements of the Earth’s outgoing radiation in the FIR for the first time and the measurements underpinning the water vapour work in this thesis were undertaken in support of the FORUM mission.

The remainder of the introduction of this thesis will cover the general concepts underlying the Earth’s radiation budget, radiative transfer through the Earth’s atmosphere and the application

of this to remote sensing. The importance of FIR radiation for the Earth's climate will be introduced with a particular focus on atmospheric water vapour and surface emissivity. The final section will lay out the structure of the rest of the thesis.

1.1 Earth's Radiation Budget

The Earth receives energy from the sun in the form of solar photons with wavelengths from 0.1 to 4 μm . In the global decadal average, the flux of these photons at the top of the atmosphere (TOA) is 340 W m^{-2} (Andrews, 2000). Some of this flux is reflected back to space by the atmosphere, clouds and the Earth's surface. The rest is absorbed (Wild et al., 2015). The Earth and atmosphere re-emit this absorbed energy in the infrared with wavelengths between 4 and $100\mu\text{m}$ (Andrews, 2000).

Figure 1.1 shows a diagram of the Earth's globally averaged radiation budget. The Intergovernmental Panel on Climate Change (IPCC) estimate that during the period 2006–2018 there was imbalance of incoming and outgoing flux of $0.79 [0.52 \text{ to } 1.06] \text{ W m}^{-2}$ (Forster et al., 2021). This imbalance has increased over time, for example the average value between 1971 and 2018 was $0.57 [0.43 \text{ to } 0.72] \text{ W m}^{-2}$. This deficit of outgoing radiation, caused by human activity (Arias et al., 2021), is a fundamental driver of global climate change.

The values in figure 1.1 are averaged globally and temporally and integrated spectrally. However the processes illustrated in figure 1.1, such as absorption and reflection of radiation in the atmosphere, exhibit complex spectral structures. Of particular interest to this thesis is the way in which the spectral structure of the outgoing longwave radiation (OLR) at TOA is influenced by the composition and properties of the Earth's surface and atmosphere.

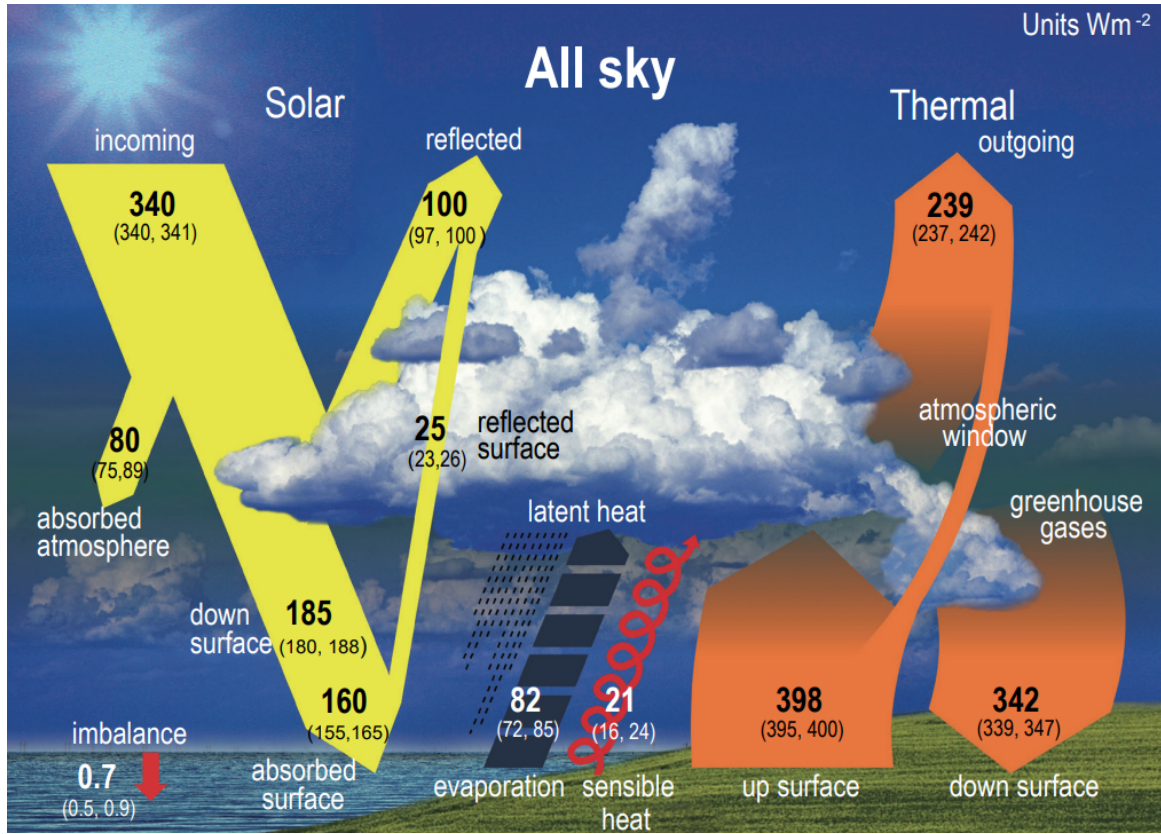


Figure 1.1: Diagram showing the globally averaged components of the Earth's energy budget for the early 21st century climate. Units are W m^{-2} with upper and lower limits in parentheses, from Forster et al. (2021) adapted from Wild et al. (2015).

Figure 1.2 shows simulations of the OLR for a typical tropical and polar winter scene in units of irradiance ($\text{W m}^{-2} \text{ cm}$). The fine structure seen in these spectra is due to some of the outgoing radiation from the Earth's surface being absorbed or scattered by atmospheric molecules. This causes the molecules to vibrate, rotate, translate or a combination of the three. As these energy transitions are quantized, the atmosphere can only absorb or emit radiation at certain energies. Photons can also cause electronic transitions in molecules but this happens at energies greater than that of thermal photons. The energy of a photon is proportional to its frequency. The frequency f of a photon can be measured in wavenumbers, ν , with units of cm^{-1} defined as

$$\nu = \frac{1}{\lambda} = \frac{f}{c}, \quad (1.1)$$

where λ is the wavelength and c is the speed of light.

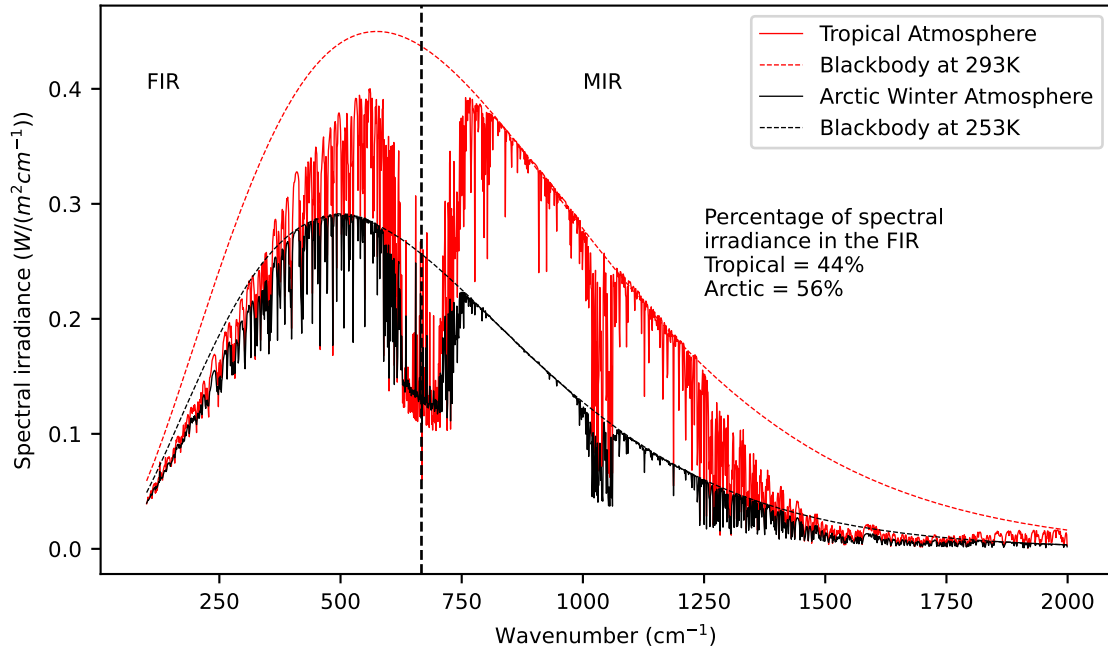


Figure 1.2: Illustrative example of the outgoing longwave flux calculated by the author using LBLRTM for a tropical (red) and arctic winter (black) atmosphere. Blackbody curves at the temperatures of the tropical and arctic surfaces are shown with dashed lines.

1.2 Radiative Transfer

The energy transferred by radiation passing through a unit area, in a specific direction, per unit time, at a specific wavenumber, is the radiance. This is commonly measured in units of $\text{W m}^{-2} \text{sr}^{-1} \text{cm}$.

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

$$dL_\nu = -k_\nu(s)\rho(s)L_\nu(s)ds, \quad (1.2)$$

where k_ν is the extinction coefficient which is a measure of how well the radiation is absorbed and

scattered by the medium at a particular frequency and ρ is the density of the medium (Andrews, 2000; Liou, 2002). If the medium is also emitting photons then an additional source term $k_\nu \rho J_\nu ds$ must be added to the right hand side of equation 1.2 to give the radiative transfer equation

$$\frac{dL_\nu}{ds} = -k_\nu \rho (L_\nu - J_\nu). \quad (1.3)$$

The optical depth τ_ν , which is a measure of how opaque the atmosphere is over a particular path between s_0 and s at a particular frequency, is given by

$$\tau_\nu = \int_{s_0}^s k_\nu \rho(s') ds' \quad (1.4)$$

and the transmittance, $\mathcal{T}_\nu$, which is the fraction of the initial radiation that is transmitted over that path is given by

$$\mathcal{T}_\nu = e^{-\tau_\nu}. \quad (1.5)$$

The definition of optical depth can be used to rewrite the radiative transfer equation (equation 1.3) to give

$$\frac{dL_\nu}{d\tau_\nu} = J_\nu - L_\nu. \quad (1.6)$$

This can then be integrated to find the spectral radiance after the radiation has passed through an optical depth τ_ν

$$L_\nu(\tau_\nu) = L_{\nu 0}e^{-\tau_\nu} + \int_0^{\tau_\nu} J_\nu(\tau')e^{-(\tau_\nu-\tau')}d\tau'. \quad (1.7)$$

The first term in equation 1.7 shows that the intensity of the radiation from the starting point, $L_{\nu 0}$ decays exponentially with respect to optical depth as it passes through the medium. If both k_ν and ρ are constant this means that the intensity of the radiation from the source decreases exponentially with distance. The second term of the equation states that the radiation is then supplemented by contributions from the radiation emitted by each element along the path also attenuated exponentially by the remaining optical depth that it must traverse (Andrews, 2000).

This can be re-written in units of distance and in terms of the transmittance to give

$$L_\nu(s_0) = L_\nu(s_0)\mathcal{T}_\nu(s_0, s) + \int_{s_0}^s J_\nu(s') \left(\frac{d\mathcal{T}_\nu(s', s)}{ds'} \right) ds'. \quad (1.8)$$

Figure 1.3 shows the transmittance through Earth's atmosphere of six species that absorb strongly in the infrared. The total spectrum is shown in the bottom panel. These data are for clear sky conditions. At wavenumbers below 600 cm^{-1} the rotational band of water vapour dominates the absorption. Centered at 667 cm^{-1} is the strong CO_2 absorption band caused by the ν_2 symmetric vibrational mode. Above this region is the atmospheric window from 800 to 1250 cm^{-1} , where the atmosphere is much more transparent, although there is some absorption particularly by ozone and CO_2 . At larger wavenumbers there is absorption by the rotation-vibration modes of water vapour and a further CO_2 vibration band (Andrews, 2000). The fingerprints of this absorption can be seen in the example OLR spectra in figure 1.2 indicating that, by measuring the OLR, information can be obtained about the underlying atmosphere. In the regions with a high transmittance, the OLR is comprised almost entirely of radiation

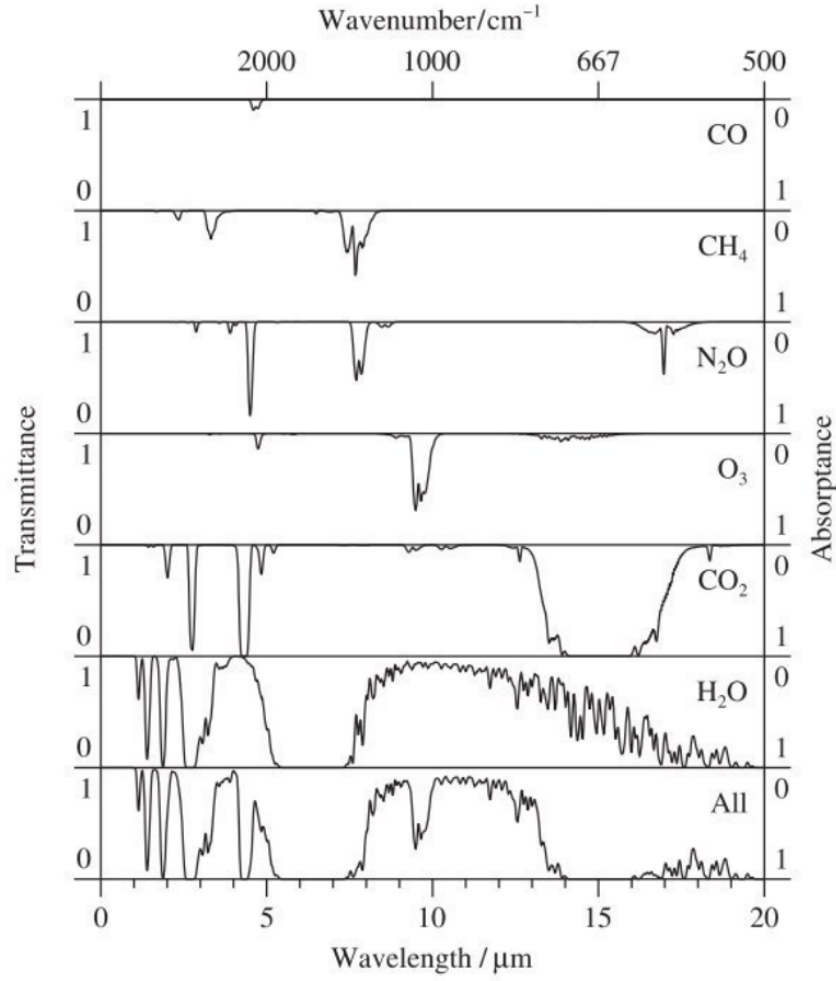


Figure 1.3: Transmittance through the Earth's atmosphere for 6 species and the total atmospheric transmission. From Andrews (2000).

emitted from the Earth's surface. In the case of figure 1.2 the Earth's surface is assumed to emit like a blackbody. A blackbody is a perfectly absorbing and emitting surface. The spectrally resolved radiance from a blackbody is a function only of temperature and is described by the Planck function,

$$B(\nu, T) = \frac{2h\nu^3}{c^2(e^{\frac{h\nu}{kT}} - 1)}, \quad (1.9)$$

where T is the temperature, h is Planck's constant, and k is Boltzmann's constant. The measured spectral radiance from a source can be converted to a brightness temperature spectrum by inverting the Planck function. At any wavenumber within the spectrum this brightness temperature is the temperature the source would have if it were emitting as a blackbody.

In reality the Earth's surface doesn't emit as a perfect black body. It is therefore useful to define the emissivity, ϵ , of a surface which is the ratio of its spectral radiance to that of a blackbody at the same temperature. The emissivity of a surface depends on multiple factors including the frequency of the radiation, viewing angle and properties of the material such as temperature or grain size (Li et al., 2013). The emissivity of a surface is one minus the reflectivity, in the case where there is no transmission through a material and the material is in thermodynamic equilibrium. In the case of a flat, homogeneous surface, such as still water or smooth ice, the reflectivity of that surface is governed by Fresnel equations and is dependent on the complex refractive index (Newman et al., 2005). The reflectivity varies with viewing angle due to Snell's law, therefore the emissivity of the surface varies with viewing angle. This angular variation also means that the emissivity of a rough surface such as snow or sand depends on surface roughness or grain size, as a rough surface can be represented by flat surfaces at a variety of viewing angles (Li et al., 2013). The surface emissivity is also related to temperature via the complex refractive index. Changing the temperature of a material changes its density and therefore its complex refractive index (e.g. Liu & Daum, 2008).

Many radiative transfer models have been developed to computationally calculate the transfer of radiation through an atmosphere. These models include line-by-line radiative transfer codes which calculate radiance at each spectral line and codes which make certain assumptions that can dramatically speed up the calculation process. This thesis makes use of the Line-By-Line Radiative Transfer Model (LBLRTM) (Clough et al., 2005; Clough et al., 1992). LBLRTM is appropriate to use for infrared radiative transfer calculations in clear-sky conditions however it does not calculate the scattering required for the simulation of cloudy scenes or the interactions between shortwave radiation and atmospheric gasses and aerosols. LBLRTM uses spectroscopic line parameters from the High-resolution Transmission Molecular Absorp-

tion (HITRAN) database (Gordon et al., 2022) and also incorporates the MT_CKD continuum model (Mlawer et al., 2012).

Continuum absorption is absorption of radiation by molecules away from the centre of discrete spectral lines. The MT_CKD model includes self- and foreign-broadened water vapour continua and continua for CO₂, O₂, N₂ and O₃. The self-broadened water vapour continuum is strongest between 760 and 1300 cm⁻¹ and at lower altitudes in the atmosphere where the water vapour density is high. On the other hand the foreign-broadened water vapour continuum is dominant in regions with strong water vapour absorption such as 50 to 300 cm⁻¹ and 1400 to 1800 cm⁻¹. It is also more important in cooler regions of the atmosphere such as the upper troposphere or Arctic environments. In the FIR the foreign broadened continuum acts to increase the opacity between the sharp water vapour lines (Clough et al., 1992). The precise causes of this absorption are not entirely known however the water vapour continuum may be linked to the interaction in between water molecules or between water molecules and other atoms in the atmosphere. The strength of the continuum absorption must be determined semi-empirically by comparison between radiative transfer calculations and measurements (e.g. Green et al., 2012; Mlawer et al., 2019). LBLRTM and the underlying databases have been extensively validated and improved by comparison with high resolution measurements (e.g. Mlawer et al., 2019; Turner et al., 2004). LBLRTM also shows good agreement with other radiative transfer codes (Kratz et al., 2005) and is considered a gold standard against which other radiative transfer codes can be compared (Oreopoulos et al., 2012).

LBLRTM is run by specifying the settings and input parameters using an input file which is read by the model when it runs. The user can specify the atmospheric profile including temperature and gas concentrations. The levels used for calculation can either be manually specified or generated automatically when the code is run. If run in vertical mode, boundary

conditions including temperature, emissivity and reflectivity can be specified. There are also six built in atmospheric profiles representing different atmospheric conditions. These profiles can be used in their entirety or used only for certain species, as specified by the user. LBLRTM can output radiances, the transmission through the full atmosphere or the optical depth of each atmospheric layer.

1.3 Remote Sensing

Remote sensing of the Earth, as opposed to in-situ measurement, involves measuring emitted or scattered radiation and using these measurements to infer some physical parameter of the Earth, its ocean or atmosphere. This can be done from the ground, from an airborne platform such as balloon or aircraft or from satellite. There are many different instruments and techniques depending on the particular parameter that is being retrieved. These instruments can be divided into active instruments which generate radiation, for example microwaves in the case of radar, and passive instruments which measure radiation that is emitted naturally (Andrews, 2000; Liou, 2002).

The work in this thesis is primarily focused on the measurement of atmospheric water vapour profiles and surface emissivity using emitted infrared radiation, therefore the remainder of this section will introduce the theory underpinning this technique.

Equation 1.7 can be adapted to the situation of a passive satellite instrument viewing the Earth directly below it (nadir viewing) to give

$$L_{\nu}(\text{TOA}) = \epsilon_{\nu} B_{\nu}(T_s) \mathcal{T}_{\nu}(0, \text{TOA}) + \int_0^{\text{TOA}} B_{\nu}(T(z)) \frac{d\mathcal{T}(z, \text{TOA})}{dz} dz. \quad (1.10)$$

Here the emission from the surface is represented by $\epsilon_\nu B_\nu(T_s) \mathcal{T}_\nu(0, \text{TOA})$ where $B_\nu(T_s)$ is the Planck function at the temperature of the surface, T_s , ϵ_ν is the emissivity and $\mathcal{T}_\nu(0, \text{TOA})$ the transmission between the surface and the top of the atmosphere. The second half of equation 1.10 represents the atmospheric emission, this assumes that the atmosphere is in local thermodynamic equilibrium and that there is no scattering of photons into the path. Therefore the source term can be written as $B_\nu(T(z))$ which is the Planck function at the atmospheric temperature at height z . $\frac{dT(z, \text{TOA})}{dz}$ is the derivative of the transmittance to space. This quantity is known as the weighting function. The value of the weighting function at a particular height represents the contribution of that height to the radiance received at the observation level. (Andrews, 2000).

Different spectral regions of the OLR can be used to sense different parameters (Houghton, 2002). For example regions with high transmission such as the atmospheric window between 800 and 1250 cm^{-1} (8-13 μm) are used to determine the sea and land surface temperature (e.g. Freitas et al., 2013) and the surface emissivity.

The atmospheric temperature profile can be retrieved using the second half of equation 1.10. This term is dependent on the vertical distribution of absorbing/emitting gases and the temperature profile of the atmospheric column below the satellite. It can be shown that the majority of upwelling radiation received by an instrument at the top of the atmosphere was emitted at unit optical depth, as measured from the top of the atmosphere. Therefore in the absence of clouds the temperature of different layers of the atmosphere can be determined by measuring the OLR in frequency bands with different absorptivities. CO_2 and O_2 are both abundant and well mixed throughout the atmosphere below the altitude of around 90 km and have absorption bands at convenient frequencies, CO_2 at 667 cm^{-1} (15 μm) and 2300 cm^{-1} (4.5 μm), and O_2 at 5 mm, therefore these regions of the OLR are commonly used for atmospheric temperature

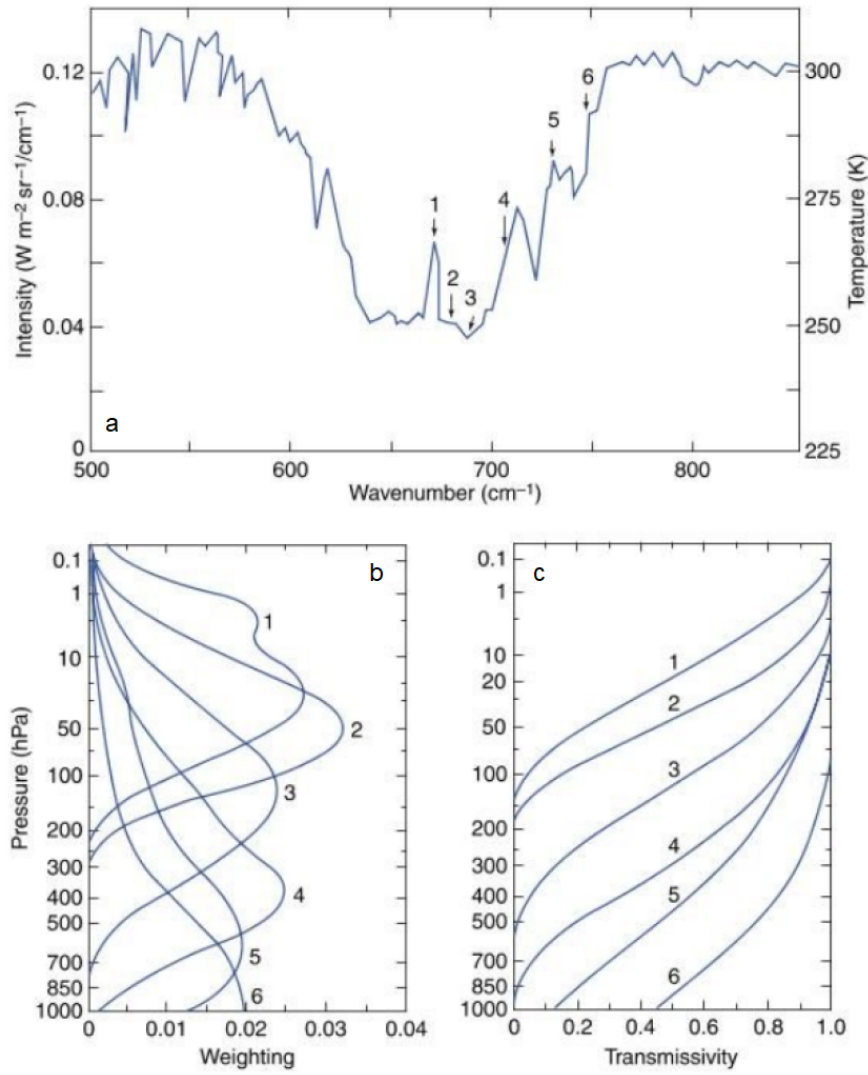


Figure 1.4: (a) Detail of the OLR around the 667 cm^{-1} CO_2 band with arrows indicating the location of six wavenumbers used in the calculation of the weighting functions shown in (b). (c) the transmissivity of the atmosphere as a function of height for the six wavenumbers defined in (a). From Wallace and Hobbs (2006) adapted from Liou (2002).

retrieval (Houghton, 2002).

Figure 1.4a shows the measured radiance at the top of the atmosphere around the 667 cm^{-1} CO_2 absorption band. Six wavenumber bands with different absorptivities are indicated. Figure 1.4b shows the weighting functions for these wavenumbers as a function of pressure, which is an alternative measure of height in the atmosphere. For example, in figure 1.4b the second weighting function peaks at around 50 hPa indicating that the radiance at 678 cm^{-1} is most sensitive to the temperature at this altitude. By sampling a range of wavenumbers with

different weighting functions the temperature of different atmospheric layers can be retrieved. Once the temperature of the atmospheric profile is known then profiles of the concentration of other non-uniformly mixed trace gases can be retrieved (e.g. Turquety et al., 2004). The mathematical basis for undertaking retrievals will be outlined in section 2.2.

1.4 The Far-infrared

The FIR consists of wavenumbers between 10 and 667 cm^{-1} (between 1000 and $15\text{ }\mu\text{m}$) accounting for $\sim 50\%$ of outgoing longwave radiation globally under all-sky conditions, see figure 1.2. This percentage is generally higher over the colder polar regions due to colder temperatures shifting the peak of the blackbody emission curve to lower wavenumbers. In the absence of clouds the outgoing spectrum in the FIR region is dominated by the rotational band of water vapour, to the extent that the lower atmosphere is generally opaque in this region. The outgoing radiation in the FIR is also sensitive to the presence of thin ice clouds. In the polar regions, the typically drier, colder atmosphere means that some FIR radiation emitted from the surface can also escape directly to space (Feldman et al., 2014).

1.4.1 Far-infrared Missions and Instruments

Satellite Instruments

The spectrally resolved outgoing FIR spectrum has not yet been systematically measured from satellite. However the recent development of uncooled detectors sensitive to FIR radiation (Palchetti et al., 2005) mean that this is now feasible and three FIR missions are currently in development.

Firstly, the European Space Agency (ESA) has selected the Far-infrared Outgoing Radiation Understanding and Monitoring (FORUM) mission as its 9th Earth Explorer mission. FORUM is scheduled to launch in 2027. Its primary instrument will be a Fourier transform spectrometer with a spectral range of 100 to 1600 cm^{-1} (100 to 6.25 μm) and a spectral resolution of 0.5 cm^{-1} (ESA, 2019). As well as providing a unique global dataset of spectrally resolved FIR radiance measurements from space, the primary objectives of the FORUM mission are to investigate spectroscopy in the FIR, provide evaluation for the radiative processes in climate models and relate observed radiative signals to atmospheric and surface variability, particularly atmospheric water vapour, clouds, greenhouse gases and surface emissivity. Secondary objectives for the FORUM mission include the retrieval of water vapour profiles, ice cloud properties and, in favourable conditions, retrieval of surface emissivity (ESA, 2019; Palchetti, Brindley, et al., 2020). The FORUM mission is of particular interest to this thesis as the work described in chapter 4 was completed in direct support of the FORUM mission.

Secondly, NASA’s Earth Venture Instrument (EVI) the Polar Radiant Energy in the Far Infrared Experiment (PREFIRE) mission consists of two small CubeSats each carrying a spectrometer with 64 spectral channels and a spectral range of 5 to 54 μm (2000 - 185 cm^{-1}) at 0.86 μm resolution. The PREFIRE mission is particularly targeted at measuring the FIR signatures of atmospheric and surface processes in the Arctic and Antarctic (L’Ecuyer et al., 2021). While the EVI call provides funding for the instrument build, the launch costs are not covered meaning PREFIRE must piggyback on a suitable scheduled launch opportunity. At present, the precise launch date is not confirmed. Finally, the Thin Ice Clouds in the Far Infrared Experiment (TICFIRE) mission, sponsored by the Canadian Space Agency, has been provisionally selected as part of NASA’s Atmosphere Observing System decadal survey mission (Blanchet et al., 2011).

Ground, Aircraft and Balloon Instruments

Very few instruments are capable of making spectrally resolved measurements across the FIR (Brindley, 2020; Harries et al., 2008). There is therefore a lack of FIR measurements, for example there have only been a handful of airborne campaigns during which spectrally resolved or narrow-band measurements of FIR radiance were made. The list below introduces the FIR instruments mentioned in this thesis.

TAFTS. The Tropospheric Airborne Fourier Transform Spectrometer (TAFTS) (Canas et al., 1997) was designed and built at Imperial College London. It is the only instrument currently capable of measuring spectrally resolved FIR radiances from aircraft. TAFTS has been deployed to make both airborne and ground based measurements. These include studies of surface emissivity (Bellisario et al., 2017; J. E. Murray et al., 2020), clear sky (Cox et al., 2007), cirrus clouds (Bantges et al., 2020; Cox et al., 2010) and the water vapour continuum (Fox et al., 2015; Green et al., 2012). TAFTS is described in more detail in section 3.1.1 and radiance measurements made using TAFTS are analysed in chapter 4.

REFIR—PAD. The Radiation Explorer in the Far InfraRed—Prototype of Applications and Development (REFIR—PAD) instrument is designed to make balloon borne and ground based measurements (Bianchini et al., 2019; Bianchini & Palchetti, 2008). It is currently installed at Concordia Station in Antarctica where it has been making near continuous measurements of downwelling radiance for almost 10 years (Palchetti et al., 2015).

FIRMOS. Following on from the design of REFIR—PAD, the Far-Infrared Radiation Mobile Observation System (FIRMOS) has been developed with support from ESA. FIRMOS

has recently been deployed to undertake a ground based field campaign in support of the FORUM mission (Palchetti, Barucci, et al., 2020).

ARI prototype. The Absolute Radiance Interferometer (ARI) prototype instrument was built to support the infrared portion of the NASA Climate Absolute Radiance and Refractivity Observatory (CLARREO) (Taylor et al., 2020). The ARI prototype instrument at the University of Wisconsin is currently configured for lab based measurements (Borbas et al., 2021).

1.4.2 The Role of Water Vapour in the Far-infrared

Water vapour makes up roughly 3% of the Earth's atmosphere however its concentration is highly variable both spatially and temporally (Andrews, 2000). As shown in figure 1.3 it absorbs strongly across the MIR and FIR so is a potent greenhouse gas. The concentration of water vapour in the atmosphere is not directly affected by human activity however the water vapour feedback is an important positive feedback in the climate system. A climate feedback is a process by which an initial change in the climate system, such as an increase in CO₂ concentration, causes a change in surface temperature which affects further secondary variables which then further change the surface temperature. The concentration of water vapour in the atmosphere is related to temperature via the Clausius-Clapeyron relation, therefore an increase in temperature causes an increase in the concentration of water vapour in the atmosphere. As water vapour is itself a greenhouse gas, this leads to further warming of the climate system (Forster et al., 2021).

The strength of the water vapour feedback is not uniform across layers of the atmosphere (Held & Soden, 2000) with enhanced sensitivity seen in the upper troposphere and lower strato-

sphere (Dessler et al., 2008; Riese et al., 2012). Accurately measuring the concentration of upper tropospheric water vapour is also important for quantifying the Earth’s greenhouse effect (Allan et al., 2002; Brindley & Harries, 1998) and understanding the atmospheric circulation (Pierrehumbert & Roca, 1998), including exchange mechanisms between the upper-troposphere and lower stratosphere (Sherwood et al., 2010; Vogel et al., 2014).

Measurements of upper tropospheric and lower stratospheric water vapour are made by radiosonde, aircraft-based sensors and a wide range of satellite instruments operating across the electromagnetic spectrum. However these current measurement techniques can have shortcomings over the upper tropospheric and lower stratospheric altitude range. For example, current water vapour retrievals from MIR hyperspectral sounders can underestimate humidity extremes and have limited sensitivity to this altitude range (Chou et al., 2009; Fetzer et al., 2008; Trent et al., 2019). Narrow-band nadir sounding instruments also have low vertical resolution around the tropopause and can suffer from clear-sky sampling biases as they cannot produce retrievals in the presence of clouds (Chung et al., 2014; John et al., 2011).

In-situ measurements, such as those from the Global Climate Observing System Reference Upper Air Network (GRUAN) radiosondes (Dirksen et al., 2014) or from instruments on commercial passenger aircraft as part of the In-service Aircraft for a Global Observing System project (Petzold et al., 2015) are restricted in their spatial coverage due to the practicalities of these in-situ measurements. Despite the work put into understanding and homogenising global water vapour data sets (McCarthy et al., 2009; McCarthy et al., 2008; Miloshevich et al., 2006; Shi & Bates, 2011), best-case uncertainties from the GRUAN radiosondes are still of the order 5%, reaching 15% near the tropopause (Dirksen et al., 2014).

Therefore there is a need for satellite measurements with good accuracy, high vertical resolution

and sensitivity to water vapour in the upper troposphere and lower stratosphere. Modelling studies indicate that some improvement may be achieved using information from hyperspectral measurements in the FIR (Clough et al., 1992; Harries et al., 2008; Merrelli & Turner, 2012; Mertens, 2002; Ridolfi et al., 2020; Shahabadi & Huang, 2014). Analysis of aircraft based measurements described in chapter 4 of this thesis and Warwick et al. (2022) have found that this improvement is also seen using real measurements.

High resolution, high accuracy measurements of the OLR in the FIR such as those made by FORUM also have the potential to verify and improve models of the spectroscopy of water vapour both by allowing measurement of spectral lines at temperatures and pressures that are not feasible in the laboratory or using ground based spectrometers and by improving estimates of the water vapour continuum coefficients (ESA, 2019). The most recently published set of coefficients for the water vapour continuum produced by Mlawer et al. (2019) using measurements from the Radiative Heating in Underexplored Bands Campaigns-II campaign still have uncertainties of up to 30% in the FIR demonstrating the need for further validation and improvement of the spectroscopy in this region.

1.4.3 Far-infrared Surface Emissivity

Emissivity is a spectrally varying property of a surface. It can depend on many properties of the surface such as its composition, roughness, temperature, grain size and grain shape. The emissivity is also dependent on the viewing angle (Li et al., 2013). Accurate knowledge of the Earth's surface emissivity is vital for our understanding of the surface energy budget as it affects the longwave flux at the Earth's surface. This then affects the amount of net surface radiation which governs the exchanges of heat and water at the surface (Liang et al., 2019). Despite this

importance, simple parameterisations or constant values of emissivity are used in most general circulation models and land surface models (Liang et al., 2019). Land surface emissivity can be retrieved from space however this is not straightforward as it involves separating the effects of the land surface temperature and land surface emissivity (Li et al., 2013). Conversely, accurate knowledge of the land surface emissivity in the atmospheric window region (800 to 1250 cm^{-1} , 8 to 13 μm) is required for accurate retrievals of land surface temperature from satellite (Jiménez-Muñoz & Sobrino, 2003).

There are several libraries of spectrally resolved emissivity values for a range of surface types, for example the ECOsystem and Spaceborne Thermal Radiometer Experiment on Space Station (ECOSTRESS) spectral library which is comprised of laboratory measurements of over 3000 material samples including soil, vegetation, water, man-made surfaces and lunar materials (Meerdink et al., 2019). The ECOSTRESS spectral library builds upon the Advanced Spaceborne Thermal Emission Reflection Radiometer (ASTER) spectral library (Baldrige et al., 2009). It should also be noted that the laboratories which make emissivity measurements differ in their measurement set-ups and protocols and a round-robin study by Langsdale et al. (2021) found differences of up to 3% for measurements of distilled water in the MIR and larger differences for less emissive surfaces.

The lack of emissivity measurements at wavenumbers below 667 cm^{-1} (above 15 μm) means that these libraries do not extend into the FIR. Very few emissivity measurements exist in the FIR and most of these are of snow and ice surfaces. These include two sets of measurements from ground based instruments and a set of airborne measurements from TAFTS. One set of ground based measurements was carried out using the FIRMOS instrument as a small part of a large winter campaign in support of the FORUM mission at the Zugspitze Observatory in the German Alps (Palchetti, Barucci, et al., 2020). Measurements of surface emissivity were made

for several snow types and were accompanied by characterisation of the snow surfaces. The results show that the emissivity varies with the snow properties. This is expected given that the measured emissivity of snow in the MIR varies with snow properties (Hori et al., 2006) and modeling indicates that this also occurs in the FIR (Chen et al., 2014). However the viewing geometry of FIRMOS made these measurements difficult to undertake (Palchetti, Barucci, et al., 2020). Measurements were also undertaken of ice, pine needles and sand by Borbas et al. (2021) using the ARI. Finally, retrievals of snow and ice emissivity were carried out using airborne measurements made by TAFTS. This was done using data from both a low altitude flight over the Greenland ice sheet (Bellisario et al., 2017) and using data from a high altitude flight over roughly the same area (J. E. Murray et al., 2020). The results from the low and high altitude flights agreed within the measurement uncertainty and demonstrate the potential for measurement of FIR snow and ice emissivity from a satellite platform. However, due to the nature of the flight campaign it was not possible to characterise the snow and ice measured. A common theme among these sets of measurements is that fact that the emissivity retrievals become much more challenging in the FIR compared to MIR. This will be discussed further in chapters 5 and 6. Despite these initial emissivity measurements, there is still a clear need for further measurements of surface emissivity that extend into the FIR.

Theoretical studies in preparation for the PREFIRE and FORUM missions indicate that this gap in emissivity measurements could be partially addressed by satellite retrievals (Ben-Yami et al., 2022; Xie et al., 2022). Both studies demonstrated successful retrievals of surface emissivity in cloud-free conditions when total column water vapour was low. However both studies also found that the success of the retrievals dramatically decreased as the total column water vapour increased, suggesting emissivity retrievals are most likely to be effective over very dry regions of the globe such as the poles. This means that both FORUM and PREFIRE should be

able to provide retrievals of snow and ice-emissivity from space. Furthermore, Ben-Yami et al. (2022) also demonstrated the theoretical retrieval of the emissivity of sand and vegetation under certain conditions. In the PREFIRE study, Xie et al. (2022) also highlighted the need for good knowledge of the a-priori surface emissivity, which can be determined from in-situ measurements. Satellite retrievals of emissivity will also need validation further emphasising the need for in-situ measurements of surface emissivity in the FIR.

These theoretical studies both made use of a database of spectral emissivity values that extends into the FIR which has been developed by Huang et al. (2016). Figure 1.5 shows a selection of the emissivity values included in the database. The database was compiled by calculating theoretical emissivity values for water, ice, snow and desert surfaces. The emissivity data for vegetation were taken from the ASTER spectral library. However, as the ASTER spectral library does not extend into the FIR, the emissivity of vegetation was assumed to be constant below 667 cm^{-1} . In the MIR the emissivity values in the database were validated by comparison to retrievals from the Infrared Atmospheric Sounding Interferometer (IASI) a hyperspectral MIR interferometer carried on the MetOp satellites.

Modelling studies undertaken using the Huang emissivity dataset indicate that including spectrally resolved FIR emissivity values can have a discernible impact on the OLR (Feldman et al., 2014; Huang et al., 2016). A further study found that incorporating the spectrally resolved emissivity values from the Huang dataset into the Community Earth System Model (CESM) led to a reduction in the temporally and spatially averaged surface temperature bias from $-4.4 \pm 2.4\text{ K}$ for the unmodified version of the CESM to a mean surface temperature bias of $-0.4 \pm 1.6\text{ K}$ for the CESM with spectrally resolved FIR emissivity (Kuo et al., 2018). This bias was calculated over the period 1979-2005 as compared to the ERA-Interim skin temperature reanalysis. The reduction in bias was most striking over the Arctic during the winter with the

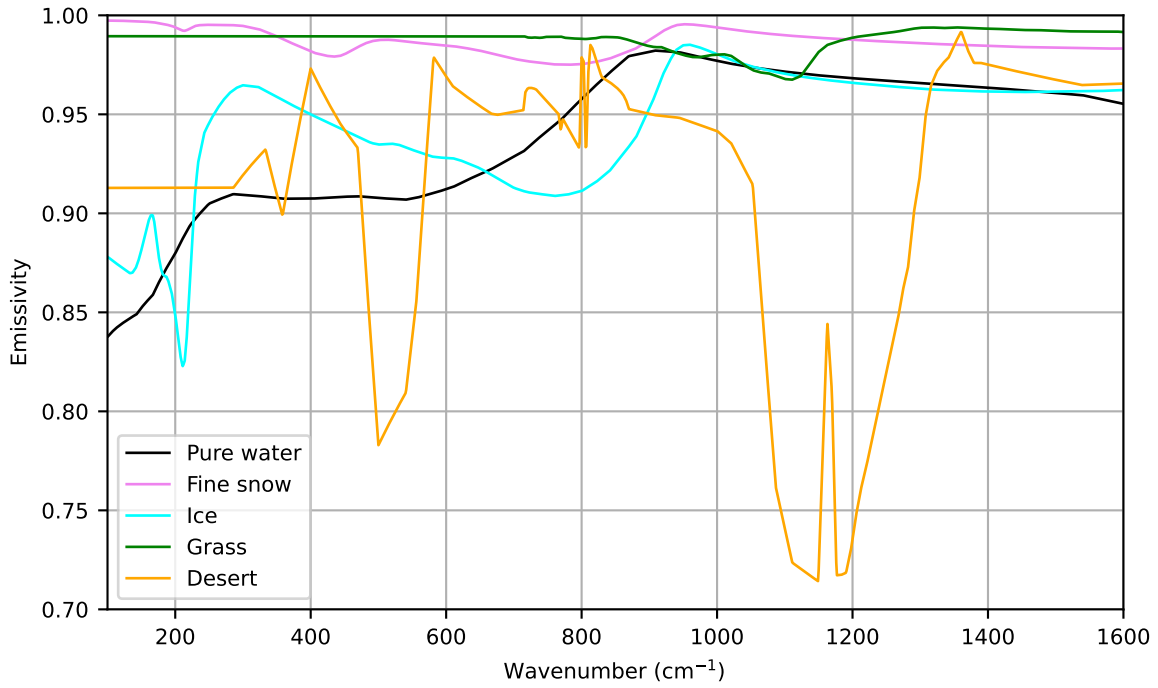


Figure 1.5: The emissivity of several surface types from the Huang et al. (2016) database.

bias reduced from -7.2 ± 0.9 K to 1.1 ± 1.2 K. The model also showed improved representation of Arctic sea ice decline. Furthermore, a separate sensitivity study undertaken using a radiative transfer code highlighted the sensitivity of the OLR to the assumed snow grain size (Chen et al., 2014). These theoretical studies further demonstrate the need for in-situ measurements of surface emissivity in the FIR particularly of snow and ice surfaces and in combination with characterisation of snow properties.

1.5 Structure of the Thesis

The work in this thesis covers two strands which both involve the use of FIR radiances to determine properties of the Earth's atmosphere and surface. These two properties are the vertical profile of atmospheric water vapour and the spectrally resolved surface emissivity.

Chapter 2 of this thesis outlines the mathematical basis of Fourier Transform spectroscopy and

the retrieval of atmospheric profiles using Optimal Estimation. Fourier Transform spectroscopy is the technique which is used to acquire the radiance spectra for both strands of this work. Optimal estimation is the technique that is used to retrieve atmospheric profiles from radiance measurements.

Chapter 3 describes the PIKNMIX-F airborne campaign and the instruments that were deployed. Jon Murray and I participated in this campaign in March 2019. These airborne measurements are a unique contribution to the small quantity of airborne FIR spectra that have been recorded. The measurements are then used in chapter 4 to investigate the sensitivity of the measured radiances to changes in atmospheric profiles, surface properties and the water vapour continuum. In collaboration with two colleagues, retrievals of the water vapour profile were also performed. These retrievals and forward modeling have been published (Warwick et al., 2022) and are the first published retrievals of atmospheric profiles performed using FIR radiance measurements from an aircraft platform.

Chapters 5 and 6 focus on the emissivity retrieval work undertaken using a new spectrometer named the Far INfrarEd Spectrometer for Surface Emissivity (FINESSE). Chapter 5 describes the method chosen to retrieve the surface emissivity and the development of FINESSE, including the calibration process, measurement uncertainty and instrument lineshape. A FINESSE simulator is developed and used to model the expected performance of the emissivity retrievals. A method for filtering the retrieved emissivity is also developed. Chapter 6 describes the first emissivity measurements made using FINESSE. These measurements are of distilled water and cover a range of surface temperatures, view angles and atmospheric conditions. The results are compared to theoretical calculations and the effect of temperature and view angle is also explored. As well as providing novel measurements of water emissivity in the FIR these first measurements from FINESSE allow us to test the operation of the instrument and our data

analysis methods before FINESSE is deployed. Finally conclusions for the work and further areas of investigation are discussed in chapter 7.

Chapter 2

Common Tools and Techniques

2.1 Fourier Transform Spectroscopy

The radiance spectra used in both strands of this thesis were measured using Fourier transform spectrometers. The spectra are acquired by performing a Fourier transform to the output of an interferometer. The design of most interferometers is based on the two-beam Michelson interferometer (Michelson & Morley, 1887), which is shown in figure 2.1, and the mathematical theory of the Michelson interferometer also applies in general to other designs of Fourier transform spectrometers. This section contains an explanation of the mathematical theory underlying the operation of an ideal Fourier Transform spectrometer and some further considerations that apply to the output of physical rather than ideal instruments, which is particularly relevant for the development of FINESSE described in chapter 5. This information is taken from textbooks written by Griffiths and de Haseth (1986) and Davis et al. (2001).

Figure 2.1 shows a schematic of a Michelson interferometer. Radiation that is incident on the aperture of the interferometer is collimated and then passed through a beam splitter. After this

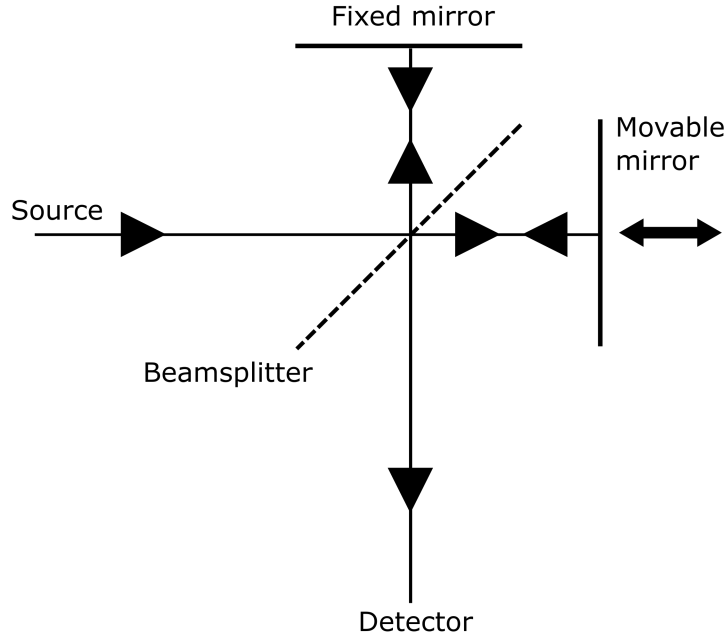


Figure 2.1: A schematic of a Michelson interferometer.

each beam of radiation is reflected by a mirror and then recombined at the same beamsplitter. One of the mirrors is movable such that the optical path difference (OPD) between the two beams of light can be varied. The output is then incident on the detector where its intensity is measured. The intensity of the light at the output varies with the OPD introduced by the movable mirror. For monochromatic light the intensity at the detector will be at a maximum when the OPD is an integer multiple of the wavelength. If the mirror is scanned continuously, the signal at the detector will vary sinusoidally as a function of OPD. The signal measured at the detector is called the interferogram. Considering just the varying part of the signal and removing any DC offset gives the mathematical expression for the interferogram $I(\delta)$,

$$I(\delta) = B(\nu) \cos(2\pi\nu\delta) \quad (2.1)$$

where δ is the OPD, ν the wavenumber of the radiation and $B(\nu)$ is the intensity of the source radiation as modified by the characteristics of the instrument, including beam splitter efficiency

and amplifier and detector response. This equation represents the Fourier transform of the input signal, modified by the instrument characteristics.

The same principle applies when a broadband source of radiation is incident on the spectrometer. In this case the energy detected is the sum of the energy detected for each frequency making up the broadband source. Therefore the expression for the interferogram is

$$I(\delta) = \int_{-\infty}^{\infty} B(\nu) \cos(2\pi\nu\delta) d\nu. \quad (2.2)$$

The input spectrum (modulated by instrument characteristics) can then be retrieved using the Fourier transform pair of equation 2.2

$$B(\nu) = \int_{-\infty}^{\infty} I(\delta) \cos(2\pi\nu\delta) d\delta. \quad (2.3)$$

In theory a spectrum of arbitrarily high resolution could be produced by scanning an arbitrarily long OPD, however this is not possible in practice and the OPD has a maximum and minimum value of $\pm\Delta$ (assumed here to be symmetrical). To account for this, the theoretical arbitrarily wide interferogram is multiplied by a truncation function $D(\delta)$ such that

$$\begin{aligned} D(\delta) &= 1 \quad \text{if} \quad -\Delta \leq \delta \leq \Delta, \\ D(\delta) &= 0 \quad \text{if} \quad \delta > |\Delta|. \end{aligned} \quad (2.4)$$

$D(\delta)$ is commonly known as a boxcar function. The spectrum is recovered by performing a

Fourier transform of the truncated interferogram

$$B(\nu) = \int_{-\infty}^{\infty} I(\delta) D(\Delta) \cos(2\pi\nu\delta) d\delta. \quad (2.5)$$

This has the effect of convolving the spectrum of arbitrary resolution with the function that is the Fourier transform of the boxcar, which is a sinc function of the form

$$f(\nu) = 2\Delta \operatorname{sinc}(2\pi\nu\delta). \quad (2.6)$$

This sinc curve has a height of 2Δ and a FWHM of $0.605/\Delta$ and is sometimes known as the “instrument line shape” (ILS) as it represents the response of the instrument to an infinitely narrow spectral line represented by a delta function. Convolving the spectrum with a sinc function produces a large “ringing” effect in the spectrum around spectral lines that are narrow compared to the ILS, therefore extra processing can be done to remove these side lobes, at the expense of the loss of some spectral resolution. This is known as apodisation. Any function that has a value of 1 at $\delta = 0$ and that decreases to zero as $|\delta|$ increases can be used as an apodisation function. Some examples of common apodisation functions include triangles and gaussians, as well as functions designed explicitly for this purpose. The ILS is then the Fourier transform of this apodisation function multiplied by a boxcar of appropriate width.

Up to this point, we have assumed that the interferogram is symmetrical and can therefore be represented by the cosine Fourier transform. However, several processes can cause the interferogram to become asymmetrical. These include an error in locating the point of zero OPD or side effects of the electronic filters designed to reduce high frequency noise. These filters can put a wavenumber dependent phase lag on each cosine component of the interferogram, meaning that the point of zero path difference depends on the frequency of the radiation. The

process of correcting for this phase lag and other phase errors to allow for the asymmetrical interferogram is known as phase correction. The phase lag or phase angle of the spectrum is usually a slowly varying function of frequency.

If the interferogram is not symmetrical due to the phase angle, as is the case for real world measurements, we must use the complex Fourier transform to retrieve the complex spectrum $B'(\nu)$

$$B'(\nu) = \int_{-\infty}^{\infty} I(\delta) e^{-i2\pi\nu\delta} d\delta. \quad (2.7)$$

The complex spectrum $B'(\nu)$ contains all the spectral information of the source but the phase angle has dispersed some of this into the imaginary plane. The true spectrum $B(\nu)$, which is a real function without noise nonlinearities caused by phase angle, can be found from the complex spectrum using

$$B(\nu) = \text{Re}(\nu) \cos(\theta_\nu) + \text{Im}(\nu) \sin(\theta_\nu) \quad (2.8)$$

where $\text{Re}(\nu)$ and $\text{Im}(\nu)$ are the real and imaginary parts of the complex spectrum $B'(\nu)$, and θ_ν is the phase angle as a function of wavenumber. This correction assumes that the phase angle is a slowly varying function of wavenumber. The phase angle can be calculated using the Mertz method (Mertz, 1965). In this method a truncated interferogram (usually of 128, 256 or 512 points) is collected around the centre burst of the interferogram. A Fourier transform is performed on this truncated interferogram, and the phase angle is calculated using

$$\theta_\nu = \arctan \left(\frac{\text{Im}(\nu)}{\text{Re}(\nu)} \right). \quad (2.9)$$

The phase angle is then interpolated to the resolution of the full spectrum and used in equation 2.8 for the phase correction.

Another factor that can cause a departure from the above theory, is the divergence of the beam of radiation as it passes through the spectrometer. The difference in distance, x , travelled by a centrally aligned ray and a ray diverging with angle α as a function of OPD is given by

$$x = \frac{2\delta}{\cos \alpha} - 2\delta \quad (2.10)$$

which can be approximated to

$$x = \delta\alpha^2 \quad (2.11)$$

if α is small. As δ increases the central ray and the diverging ray become increasingly out of phase and the contrast between light and dark fringes at the detector decreases until the two rays are completely out of phase and the fringe contrast at the detector disappears completely (Griffiths & de Haseth, 1986). This has the effect of limiting the resolution of the interferogram as no further information is added by scanning beyond this limit. Therefore, it is sometimes called self-apodisation. This beam divergence also has the effect of introducing a wavenumber dependent frequency shift in the wavenumber scale recorded by the interferometer due to the different effective path differences seen by the central and extreme rays. Additionally, many designs of Fourier transform spectrometers contain a metrology laser which governs the sampling of the interferogram by measuring the position of the scanning mirror. The metrology laser must be slightly offset from the infrared optical path and any misalignment between these two systems can also introduce a wavenumber dependent frequency shift. This shift can be

determined by a comparison between the measured spectrum and a reference spectrum, such as one calculated using a radiative transfer code. Once determined the wavenumber shift should remain consistent unless the alignment of the interferometer or the average angle of the beam passing through the interferometer is changed.

Finally, to determine the absolute radiance measured by the spectrometer, the response function of the instrument must be determined. A single measured spectrum $B(\nu)$ can be related to the absolute radiance signal $L(\nu)$ via the equation

$$B(\nu) = R(\nu)L(\nu) + C(\nu), \quad (2.12)$$

where $R(\nu)$ is the instrument response function and $C(\nu)$ is a background offset which can be caused by instrument self-emission or atmospheric emission. Assuming $C(\nu)$ is constant across measurements, the instrument response function can be calculated by differencing two measurements of known radiance. Usually these are a hot blackbody calibration target and a cold or ambient temperature blackbody calibration target. The instrument response function is then calculated using

$$R(\nu) = \frac{B_{HBB}(\nu) - B_{CBB}(\nu)}{L_{HBB}(\nu, T_{HBB}) - L_{CBB}(\nu, T_{CBB})} \quad (2.13)$$

where B_{HBB} and B_{CBB} are the measured signals from the hot and cold blackbodies and L_{HBB} and L_{CBB} are the blackbody radiances from the hot and cold blackbodies at temperatures T_{HBB} and T_{CBB} . This assumes that the blackbodies have a uniform temperature and an emissivity of 1.

By considering equation 2.12, we can find the calibrated scene radiance, L_{scene} , by perform-

ing another difference against the measured signal from the hot blackbody calibration target. This cancels out the background offset $C(\nu)$ and means the calibrated scene radiance can be determined using

$$L_{\text{scene}}(\nu) = \frac{B_{\text{scene}}(\nu) - B_{HBB}(\nu)}{R(\nu)} + L_{HBB}(\nu). \quad (2.14)$$

2.2 Retrieval Theory

The retrieval of temperature and water vapour profiles from the measurement of outgoing infrared radiation is a complex inverse problem that is often poorly constrained. A detailed description of the methods for solving these problems can be found in *Inverse Methods for Atmospheric Sounding* by Rodgers (2000). The following description is a basic summary based on this book with the aim of explaining the terminology which will be used in chapter 4.

The measurements made at the top of the atmosphere, $\mathbf{y}$ can be linked to parameters to be retrieved, $\mathbf{x}$ by a forward model, $\mathbf{F}(\mathbf{x})$:

$$\mathbf{y} = \mathbf{F}(\mathbf{x}) + \boldsymbol{\epsilon}, \quad (2.15)$$

where $\mathbf{y}$, $\mathbf{x}$ and $\boldsymbol{\epsilon}$ are vectors with $\boldsymbol{\epsilon}$ being the measurement error. In the case of remote sensing of temperature and water vapour using infrared radiances, $\mathbf{y}$ could be the measured spectrum with error $\boldsymbol{\epsilon}$, $\mathbf{F}(\mathbf{x})$ a radiative transfer model, and $\mathbf{x}$ the profile of temperature and water vapour concentration.

It is often convenient to linearise equation 2.15 around a reference state $\mathbf{x}_0$ to give

$$\mathbf{y} - \mathbf{F}(\mathbf{x}_0) = \frac{\partial \mathbf{F}(\mathbf{x})}{\partial \mathbf{x}}(\mathbf{x} - \mathbf{x}_0) + \boldsymbol{\epsilon} = \mathbf{K}(\mathbf{x} - \mathbf{x}_0) + \boldsymbol{\epsilon}, \quad (2.16)$$

where $\mathbf{K}$ is a matrix of derivatives and in this thesis will be referred to as the Jacobian, although it can also be called the weighting function matrix or the kernel.

A Bayesian approach can be used to solve the inverse problem. This solution gives the probability density function of the underlying atmospheric state given a particular measurement.

Bayes theorem states

$$P(\mathbf{x}|\mathbf{y}) = \frac{P(\mathbf{y}|\mathbf{x})P(\mathbf{x})}{P(\mathbf{y})}, \quad (2.17)$$

where, in the case of atmospheric retrievals:

- $P(\mathbf{x}|\mathbf{y})$ is the probability density function of the atmospheric state given a particular measurement. This is the the quantity that the retrieval is trying to obtain.
- $P(\mathbf{x})$ is the a-priori knowledge of the atmospheric state.
- $P(\mathbf{y}|\mathbf{x})$ is the information about $\mathbf{y}$ which would be obtained by a measurement of $\mathbf{x}$. This can be calculated using the forward model and knowledge of the error.
- $P(\mathbf{y})$ is usually only a normalising factor and often does not need to be calculated.

For a retrieval problem where the forward model is linear and the measurement statistics are Gaussian, $P(\mathbf{y}|\mathbf{x})$ can be expressed as:

$$-2 \ln P(\mathbf{y}|\mathbf{x}) = (\mathbf{y} - \mathbf{K}\mathbf{x})^T \mathbf{S}_{\mathbf{y}}^{-1} (\mathbf{y} - \mathbf{K}\mathbf{x}) + c_1, \quad (2.18)$$

where c_1 is a constant, $\mathbf{S}_{\mathbf{y}}$ is the covariance matrix representing the measurement error, the superscript T indicates the transpose and the superscript -1 indicates the matrix inverse. The

diagonal elements of $\mathbf{S}_y$ represent the uncorrelated uncertainty on each element of $\mathbf{y}$ and the off diagonal elements represent the correlation between elements. Also assuming Gaussian statistics, $P(\mathbf{x})$ can be written as:

$$-2 \ln P(\mathbf{x}) = (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a) + c_2, \quad (2.19)$$

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.

Substituting equations 2.18 and 2.19 into equation 2.17 gives an equation for the probability density function after the measurement:

$$-2 \ln P(\mathbf{x}|\mathbf{y}) = (\mathbf{y} - \mathbf{K}\mathbf{x})^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{K}\mathbf{x}) + (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a) + c_3. \quad (2.20)$$

This is also a Gaussian probability density function with a covariance, $\hat{\mathbf{S}}$, given by the expression

$$\hat{\mathbf{S}}^{-1} = \mathbf{K}^T \mathbf{S}_y^{-1} \mathbf{K} + \mathbf{S}_a^{-1}. \quad (2.21)$$

and with an expected value, $\hat{\mathbf{x}}$, given by

$$\hat{\mathbf{x}} = \mathbf{x}_a + (\mathbf{K}^T \mathbf{S}_y^{-1} \mathbf{K} + \mathbf{S}_a^{-1})^{-1} \mathbf{K}^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{K}\mathbf{x}_a) = \mathbf{x}_a + \mathbf{G}(\mathbf{y} - \mathbf{K}\mathbf{x}_a), \quad (2.22)$$

where $\mathbf{G}$ is the gain matrix.

When the linear assumption is not sufficient to solve the retrieval problem, equation 2.20 must be adapted. In the non-linear case where the measurement error is Gaussian and there is an a-priori estimate with Gaussian error equation 2.20 becomes

$$-2 \ln P(\mathbf{x}|\mathbf{y}) = [\mathbf{y} - \mathbf{F}(\mathbf{x})]^T \mathbf{S}_y^{-1} [\mathbf{y} - \mathbf{F}(\mathbf{x})] + [\mathbf{x} - \mathbf{x}_a]^T \mathbf{S}_a^{-1} [\mathbf{x} - \mathbf{x}_a] + c_3. \quad (2.23)$$

Additionally, in the non-linear case the expected value of $P(\mathbf{x}|\mathbf{y})$ may not be a good representation of the most likely state. Therefore the maximum probability state is found by differentiating equation 2.23 and setting it equal to zero to give

$$-\hat{\mathbf{K}}^T(\hat{\mathbf{x}}) \mathbf{S}_y^{-1} [\mathbf{y} - \mathbf{F}(\hat{\mathbf{x}})] + \mathbf{S}_a^{-1} (\hat{\mathbf{x}} - \mathbf{x}_a) = 0, \quad (2.24)$$

where $\mathbf{K}(\mathbf{x}) = \nabla_{\mathbf{x}} \mathbf{F}(\mathbf{x})$ is the Jacobian matrix. This equation must be solved numerically which can be achieved by adapting the Newton–Raphson method for finding roots of functions. For the matrix case the Newton-Raphson method is

$$\mathbf{x}_{k+1} = \mathbf{x}_k - [\nabla_{\mathbf{x}} \mathbf{g}(\mathbf{x}_k)]^{-1} \mathbf{g}(\mathbf{x}_k) \quad (2.25)$$

where $\mathbf{x}_{k+1}$ is the updated estimate of the root, $\mathbf{x}_k$ is the current estimate of the root, $\mathbf{g}$ is given by the left hand side of equation 2.24 and $\nabla_{\mathbf{x}} \mathbf{g}$ is given by

$$\nabla_{\mathbf{x}} \mathbf{g} = \mathbf{S}_a^{-1} + \mathbf{K}^T \mathbf{S}_y^{-1} \mathbf{K} - [\nabla_{\mathbf{x}} \mathbf{K}^T] \mathbf{S}_y^{-1} [\mathbf{y} - \mathbf{F}(\mathbf{x})]. \quad (2.26)$$

The term of this equation containing the derivative of the Jacobian matrix, $[\nabla_{\mathbf{x}} \mathbf{K}^T] \mathbf{S}_y^{-1} [\mathbf{y} - \mathbf{F}(\mathbf{x})]$, is expensive and complex to compute but luckily, in cases that are only moderately non-linear, this is much smaller than the other terms of the equation and can be omitted. Substituting equation 2.24 and the simplified form of equation 2.26 into equation 2.25 gives the Gauss-

Newton method:

$$\mathbf{x}_{k+1} = \mathbf{x}_k + (\mathbf{K}_k^T \mathbf{S}_y^{-1} \mathbf{K}_k + \mathbf{S}_a^{-1})^{-1} [\mathbf{K}_k^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{F}(\mathbf{x}_k)) + \mathbf{S}_a^{-1} (\mathbf{x}_a - \mathbf{x}_k)]. \quad (2.27)$$

Sometimes the Gauss-Newton method can fail to converge to a solution because the true solution is too far from the current iteration and the problem is too non-linear. In these cases an extra parameter can be inserted into the equation to give

$$\mathbf{x}_{k+1} = \mathbf{x}_k + (\mathbf{K}_k^T \mathbf{S}_y^{-1} \mathbf{K}_k + \mathbf{S}_a^{-1} + \lambda_k \mathbf{D})^{-1} [\mathbf{K}_k^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{F}(\mathbf{x}_k)) + \mathbf{S}_a^{-1} (\mathbf{x}_a - \mathbf{x}_k)], \quad (2.28)$$

where λ_k is the Levenberg-Marquardt parameter and $\mathbf{D}$ is a diagonal matrix. The Levenberg-Marquardt parameter can be adjusted at each iterative step to aid the convergence to the solution. The exact form of the parameter and the criteria for its adjustment vary between different applications and are generally found experimentally for different problems.

An important quantity for evaluating the performance of a retrieval is the number of degrees of freedom for signal (DFS). The DFS is a positive number quantifying the number of independent pieces of information added by a measurement. A higher value means more information is provided.

The DFS is given by

$$\text{DFS} = \text{trace} [(\mathbf{K}^T \mathbf{S}_y^{-1} \mathbf{K} + \mathbf{S}_a^{-1})^{-1} \mathbf{K}^T \mathbf{S}_y^{-1} \mathbf{K}] = \text{trace} (\mathbf{GK}) = \text{trace}(\mathbf{A}). \quad (2.29)$$

Here $\mathbf{A}$ is the averaging kernel matrix. It represents how the retrieval smooths out the true atmospheric state and thus gives an indication of the vertical resolution of the retrieval. The DFS is dependent on the uncertainty in the a-priori, with larger a-priori uncertainty leading to higher DFS; the uncertainty of the measurements, with higher uncertainty leading to lower DFS; and on the Jacobian, which describes how sensitive the forward model is to changes in the inputs or target variable.

This retrieval method will be used to perform the retrievals of temperature and water vapour described in chapter 4 and the DFS will be used to evaluate the retrievals.

Chapter 3

The PIKNMIX Measurement Campaign and Instrumentation

This chapter describes the PIKNMIX-F field campaign and the instrumentation and additional data sets that are used in chapter 4.

The PIKNMIX-F field campaign was an extended airborne field campaign that took place in March 2019 based out of Stornoway, Scotland. The campaign was a joint endeavour between the UK Met Office and the Facility for Airborne Atmospheric Measurements (FAAM) Airborne Laboratory. The main science goals of the campaign were defined by the Met Office however ESA provided additional funding that allowed further flight hours dedicated to supporting the FORUM mission. These dedicated FORUM flying hours included high altitude flights in clear sky conditions and overflights of cirrus clouds. Table 3.1 describes the flights from the PIKNMIX-F campaign that are of particular interest for the FORUM mission. As well as the work described in this thesis, data from these FORUM flight hours have been used in other studies to investigate the ability of the FIR to identify cirrus clouds (Magurno et al., 2020) and

to investigate water vapour spectroscopy.

Flight No.	Date	Conditions	Altitude	Sondes	Satellite Overpasses
C153	13/03/2019	Clear Sky	8500 to 9100 m	8	Suomi-NPP & AQUA AEOLUS
C158	18/03/2019	Clear Sky	10000 m	8	
C164	25/03/2019	Cirrus	9800 m	4	
C168	28/03/2019	Cirrus	10000 m	5	

Table 3.1: Flights from the PIKNMIX-F campaign in which TAFTS participated and which are of particular interest to the FORUM mission.

The work in chapter 4 of this thesis is based on flight C153. This clear-sky flight took place on 13th March 2019 and had the aim of observing co-incident nadir-viewing spectra in the FIR and MIR at high altitude from the Tropospheric Airborne Fourier Transform Spectrometer (TAFTS) and the Airborne Research Interferometer Evaluation System (ARIES) respectively (Warwick et al., 2022). There was also frequent characterisation of the underlying atmosphere from sonde profiles. Flight C153 was chosen for this clear sky analysis due to the overpass of the AQUA satellite which carries the Atmospheric Infrared Sounder (AIRS) a hyperspectral MIR instrument (Aumann et al., 2003).

Figure 3.1 shows the flight track for C153. As can be seen from the background image, which is taken from the Spinning Enhanced Visible and InfraRed Imager (SEVIRI) on Meteosat-11, conditions in the operating area were generally clear but with a small amount of low cloud visible at the southernmost point of the flight track. After transiting to over the North Sea, the aircraft performed two straight and level runs (SLRs) between 57 and 55°N shown in orange in figure 3.1. During these SLRs coincident nadir viewing measurements were made by TAFTS and ARIES. The first SLR was flown at around 8500 m in a southerly direction and the second SLR was flown in a northerly direction at around 9100 m. The SLRs were started at 1155 UTC and completed at 1255 UTC. After the SLRs the aircraft performed a spiral descent pattern, followed by a final SLR at 300 m and then returned to Stornoway.

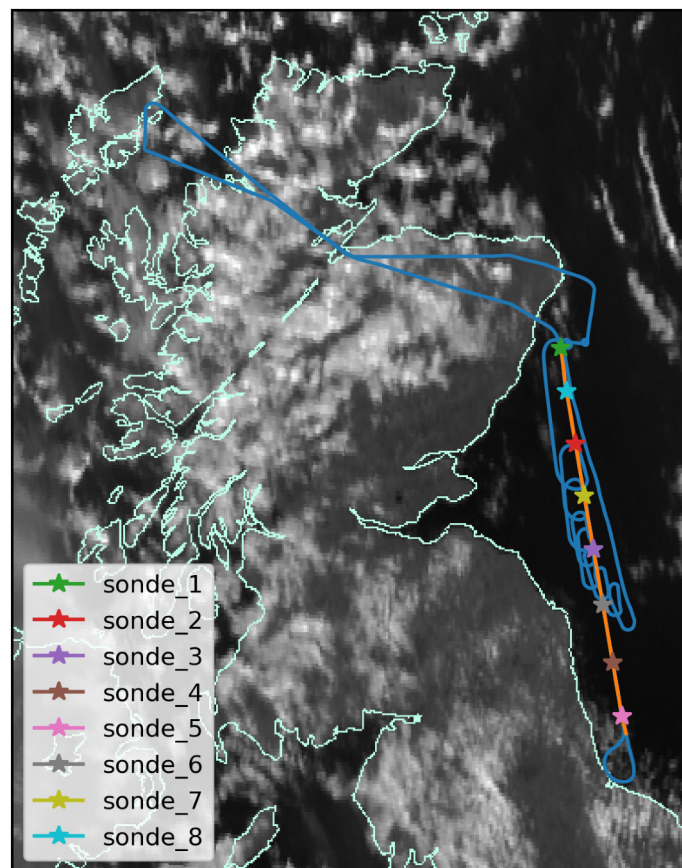


Figure 3.1: The flight track for C153. The orange portion shows the high level runs and the stars indicate the sonde release locations. The background is the high-resolution visible image from SEVIRI on Meteosat-11 at 12:30UTC. Figure from Warwick et al. (2022) produced by Stuart Fox.

3.1 Radiation Instrumentation

3.1.1 The Tropospheric Airborne Fourier Transform Spectrometer (TAFTS)

The Tropospheric Airborne Fourier Transform Spectrometer (TAFTS) is a polarizing Martin-Puplett interferometer designed and built at Imperial College for the measurement of high-resolution FIR spectra from aircraft (Canas et al., 1997). Table 3.2 summarises TAFTS's

key characteristics. It has a nominal spectral range of 80 to 600 cm^{-1} , split into a longwave (lw) channel with a range 80 to 300 cm^{-1} and a shortwave (sw) channel with a range 330 to 550 cm^{-1} . The maximum spectral resolution of TAFTS is 0.12 cm^{-1} however during the PIKNMIX-F campaign it was decided that the OPD of TAFTS should be reduced to increase the signal to noise ratio by a factor of $\sqrt{2}$ and reduce the time taken for TAFTS to perform a single scan. This also had the effect of reducing the resolution of TAFTS to 0.24 cm^{-1} .

Instrument	TAFTS	ARIES
Detectors	GeGa (lw) and SiSb (sw)	HgCdTe (lw) and InSb (sw)
Range (cm^{-1})	80 – 300 (lw) and 330 – 550 (sw)	550 – 1800 (lw) and 1700 – 3000 (sw)
Nominal Resolution (cm^{-1})	0.12	1
Scan time (s)	1.5	0.25

Table 3.2: Key characteristics of the TAFTS and ARIES interferometers.

TAFTS can be mounted in a blister on the side of the FAAM Airborne Laboratory allowing it to acquire nadir and zenith views. Figure 3.2 shows the FAAM aircraft on the ground including the location of TAFTS and ARIES. TAFTS is designed in a four-port configuration. It has four black body calibration targets, one hot and cold pair for each input port. For these measurements the hot black body targets were heated to around 50°C and the cold targets were held near ambient temperature. The temperatures of all calibration targets were monitored using platinum resistance thermometers.

The work in this thesis made use of TAFTS spectra which had been processed and calibrated by Jon Murray. The raw data recorded by TAFTS were processed using the IDL routine developed by Paul Green and described in Beeby (2011). In this routine the raw interferograms are interpolated to allow for variations in the movement of the TAFTS scanning mirror, the centreburst of each interferogram is identified and a complex Fourier transform is performed

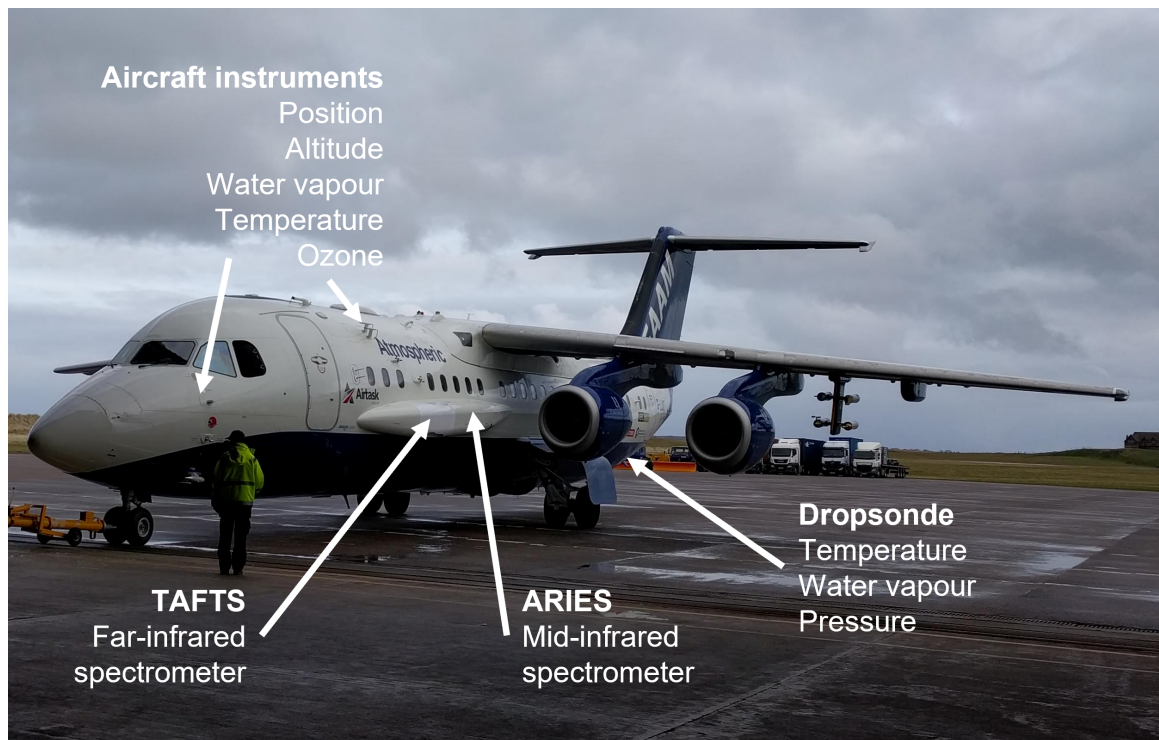


Figure 3.2: A photograph of the FAAM aircraft showing the location of the radiation and auxiliary instrumentation

to produce complex spectra. The phase angle is calculated for each complex spectrum using equation 2.8. Finally the spectra are “de-ghosted” this compensates for the effect of vibrations of the beamsplitter. The spectra were then calibrated and phase corrected using a method developed by Jon Murray which is described in J. Murray (2019). In this method the instrument self emission term is isolated by performing differences of different viewing configurations. It should be noted that this method of calibration is different to that described in Beeby (2011). These measurements were accompanied by uncertainty estimates comprising of a systematic component related to the calibration error introduced by uncertainty in the temperatures of the black body calibration targets and a random component related to random noise. This random noise is due to a combination of photon noise, which is noise caused by the inherently random timing of the photons being received and counted by the detector, and additional random noise caused by the effect of random vibrations that the instrument undergoes on the aircraft which are not removed by the “de-ghosting” procedure (Jon Murray, personal communication 2023).

The random component could be reduced by averaging spectra. For any given individual or average spectrum the total error was found by adding the systematic, and appropriately treated random uncertainty in quadrature. These data can be accessed on the CEDA archive (Facility for Airborne Atmospheric Measurements; Natural Environment Research Council; Met Office, 2019).

3.1.2 The Airborne Research Interferometer Evaluation System (ARIES)

ARIES is a Michelson-type MIR spectrometer (Wilson et al., 1999). Table 3.2 summarises its key properties. It is designed for measurements from aircraft and can sit in the blister on the side of the FAAM Airborne Laboratory adjacent to TAFTS. ARIES has a spectral resolution of 1 cm^{-1} and a spectral range of 550 to 3000 cm^{-1} . For the work in this thesis calibrated ARIES spectra were provided by Stuart Fox. These data are also available on the CEDA archive (Facility for Airborne Atmospheric Measurements; Natural Environment Research Council; Met Office, 2019). These ARIES spectra were also accompanied by errors consisting of a systematic and random component.

Figure 3.3 shows a comparison between the average of 10 seconds of data measured by TAFTS and ARIES. The spectra tend to agree in the overlap region within the quoted instrument uncertainties.

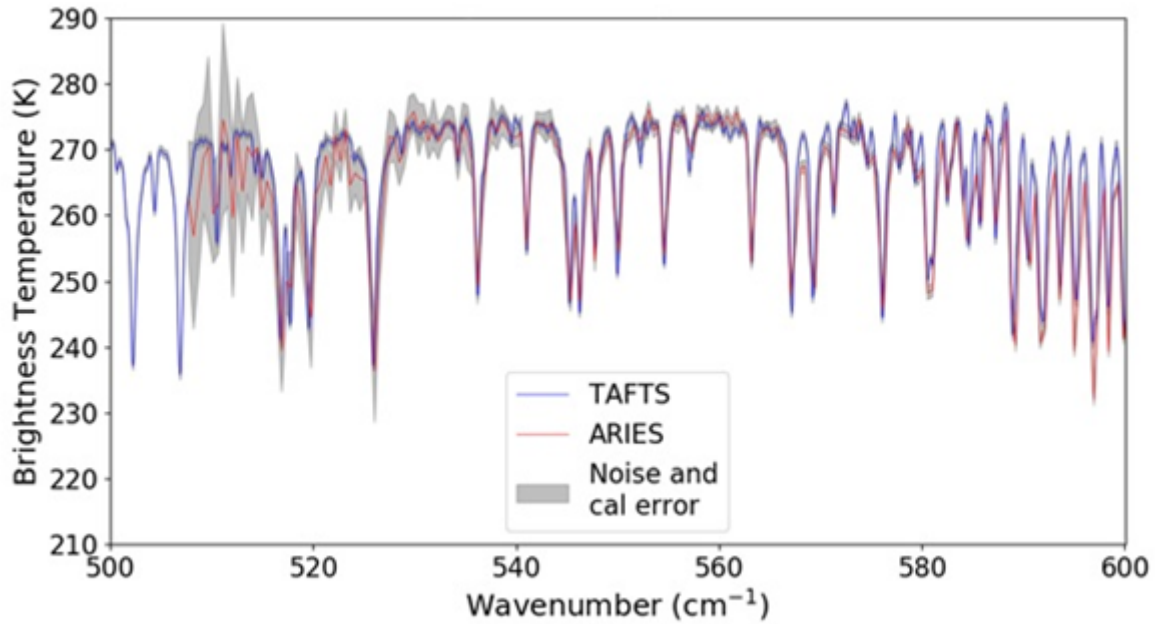


Figure 3.3: The spectra in units of brightness temperature recorded by TAFTS (blue) and ARIES (red) plotted in the wavenumber region where they overlap. The grey shading represents the random uncertainty and calibration error for each spectrum added in quadrature. These spectra are the average of 10 seconds of data taken from the middle of the second SLR.

3.2 Aircraft Data

Additional data were taken from the core instruments on the FAAM Airborne Laboratory (FAAM Instrument Team, 2019; Facility for Airborne Atmospheric Measurements; Natural Environment Research Council; Met Office, 2019). Table 3.3 summarises the instruments used to acquire these additional data.

During the high level SLRs the atmosphere below the aircraft was characterised by the release of eight Vaisala RD94 dropsondes (sondes), see figure 3.1. As they fell the sondes made measurements of position, pressure, temperature and humidity. The manufacturer states that the repeatability of the sonde temperature measurement is 0.2 K (Vaisala, 2017). The uncertainty in the humidity measurement from the Vaisala RS92 radiosonde which uses the same humidity sensor as the Vaisala RD94 dropsonde has been assessed by Miloshevich et al. (2006). They

Variable	Derivation
Position	POS AV 410 GPS-aided Inertial Navigation unit
Static pressure	Air Data Computer on G-LUXE
Static air temperature	Derived from measurements made using a Rosemount Aerospace Inc. Type 102 De-iced Total Temperature Housing fitted with an IST MiniSens PRT sensor, and the Air Data Computer on G-LUXE
Water vapour volume mixing ratio	Derived from dewpoint measurements made using a Buck Research Instruments CR2 Chilled Mirror Hygrometer with heated inlet and information from the Air Data Computer
Ozone volume mixing ratio	Core Thermo Fisher Scientific Inc. model 49i UV absorption ozone photometer (hereafter TECO 49)
Vertical temperature and water vapour volume mixing ratio profiles	Advanced Vertical Atmospheric Profiling System on the aircraft releasing Vaisala RD94 dropsondes

Table 3.3: Origin of data taken from the core instruments on board the FAAM aircraft (FAAM Instrument Team, 2019).

found the uncertainty had two components related to the production variability and sensor calibration. The uncertainty due to the calibration was found to be $\pm 5\%$ of the measured relative humidity value plus an absolute offset of $\pm 0.5\%$ and the uncertainty due to production variability was found to be $\pm 1.5\%$ of relative humidity values above 10% or $\pm 3\%$ for values below 10%.

3.3 Additional Data

At the end of the second high level SLR, the flight was overpassed by the AIRS instrument on the AQUA satellite. The level 2 products of AIRS include water vapour and temperature profiles and surface temperature derived using MIR radiance measurements. For this work the AIRS only Version 6 Level 2 temperature and water vapour retrieval products (Susskind et al., 2003) were used to provide an additional characterisation of the atmospheric state derived from

MIR radiances at the time of the aircraft observations.

The global forecast from the operational Met Office Unified Model (UM) was also used in the analysis (Walters et al., 2017). This model has a grid length of around 10 km at the latitude of the UK and 70 vertical levels. The model error is defined by the background error covariance matrix (B-matrix). This has 144 rows/columns. 140 rows/columns correspond to the correlated errors in the temperature and specific humidity for each model level, the remaining four elements represent the correlated errors in the near surface temperature, near surface humidity, surface pressure and surface skin temperature. The B-matrix is positive definite and symmetrical.

3.4 Conclusion

The measurements taken during the PIKNMIX-F campaign are a unique dataset comprising of co-located MIR and FIR spectra. These measurements comprise upwelling radiances of clear and cirrus scenes over ocean in the mid-latitudes. The measurements have a lot of potential to inform novel scientific studies. These include the development and inter-comparison of radiative transfer and retrieval codes in the FIR. The development of these codes is particularly important at the moment due to the upcoming launches of PREFIRE and FORUM. The measurements are also being used to investigate the radiative properties of cirrus clouds in the FIR and test models of the optical properties of ice clouds. The data from the clear sky flight C153 are analysed in chapter 4 with a focus on atmospheric water vapour.

Chapter 4

Retrieval of Water Vapour Profiles from PIKNMIX-F

The aim of this chapter is to investigate whether FIR radiance measurements made from aircraft during PIKNMIX-F flight C153 can be used to improve the information about the atmospheric water vapour profile and how this improvement compares to retrievals made using MIR radiance measurements. Section 4.1 discusses forward modelling of the measured radiances. The aim of this forward modelling is to investigate the sensitivity of the measured radiance to changes in atmospheric profiles, surface properties and water vapour spectroscopy. This clear sky work follows on from the previous study of Cox et al. (2007) which found that the far-infrared spectrum showed sensitivity to the atmospheric humidity profile. Since this study, changes have been made to the water vapour spectroscopy (e.g. Green et al., 2012; Rothman et al., 2013) and the TAFTS calibration process (see section 3.1.1) including an upgrade to the PRTs monitoring the temperatures of the blackbody calibration targets (Beeby, 2011, Jon Murray personal communication 2023). Section 4.2 contains retrievals carried out using measured

FIR and MIR radiances and a comparison of the information added by the FIR and MIR measurements. These are the first water vapour retrievals carried out using FIR radiance measurements from an aircraft platform. The chapter finishes with a section of conclusions.

4.1 Forward Modelling

The aim of this section is to carry out forward modelling to investigate the sensitivity of the FIR spectrum to changes in atmospheric profile, surface properties and water vapour continuum and compare these simulations to the measurements of infrared radiance described in chapter 3. It focuses on the data taken around the release of sonde 8 at the north end of the second SLR of flight C153 as this coincided with the overpass of the AIRS instrument.

Forward modelling of infrared radiance at the aircraft level was carried out using LBLRTM v12.8 (Clough et al., 2005), see section 1.2. Three main model atmospheres were used, which are summarised in Table 4.1.

Case	Atmospheric profiles	Profile Uncertainty	Surface Temperature
UM	Met Office UM	Met Office UM B-matrix	Met Office UM ($279.63 \pm 2.3\text{K}$)
Sonde	Sonde 8	Measurement uncertainty from (Vaisala, 2017) and (Miloshevich et al., 2009)	IREMIS simulation ($280.25 \pm 0.75\text{K}$)
AIRS	AIRS retrieval	AIRS retrieval	AIRS retrieval ($280.6 \pm 4.6\text{K}$)

Table 4.1: Specifications of the three model atmospheres used for forward modelling.

The first model atmosphere, UM, used temperature and humidity profiles and surface skin temperature from the T+1 hour Met Office UM at an initialisation time of 12 UTC at the closest grid point to the release of sonde 8. The aircraft was 3 km from the centre of the grid

box when the sonde was released. The diagonal elements of the B-matrix were used to quantify the uncertainty in the temperature and water vapour profiles.

The sonde case used temperature and humidity profiles measured by sonde 8 and measurements of temperature and humidity taken at the aircraft level. UM data were used to fill the 800 m gap between these two sets of measurements. The UM data did not require scaling to be consistent with the in-situ data. The uncertainty in the atmospheric profiles used in the sonde case was determined from the repeatability of the sonde temperature measurements and the uncertainty assessment of the humidity measurement described in section 3.2. The surface temperature for the sonde case was provided by Stephan Havemann and was estimated using the IREMIS emissivity model. IREMIS is used to calculate surface emissivity in the Radiative Transfer for TOVS (RTTOV) model (Saunders et al., 2017). IREMIS takes inputs of surface skin temperature, surface wind speed and sensor viewing zenith and returns an emissivity spectrum. To estimate the surface skin temperature, an initial guess of surface skin temperature, a sensor zenith angle of 0° and zonal and meridional wind speeds of 10 m s^{-1} and 3 m s^{-1} were used as inputs to IREMIS to generate an initial estimate of the surface emissivity. This initial emissivity spectrum was used as input to the line-by-line version of the Havemann-Taylor radiative transfer code (Havemann et al., 2018) to produce expected radiances at the aircraft level. These were compared to ARIES radiances between 880 cm^{-1} and 920 cm^{-1} . The surface skin temperature used as input to IREMIS was then iteratively altered which in turn altered the simulated emissivity spectrum. New expected radiances were generated until the RMS difference between the simulation and ARIES observations was minimised. Using this iterative method, Stephan Havemann found that a surface skin temperature of 280.25 K minimised the differences between the simulations and observations. No uncertainty value was provided with this estimate however subsequent analysis (see section 4.1.2) showed that

a surface temperature uncertainty of 0.75 K was consistent with the radiance uncertainty of ARIES in the 880 cm^{-1} to 920 cm^{-1} spectral region. The emissivity spectrum also generated using this iterative method was used for all simulations in this section.

Finally, the AIRS case used the temperature and humidity profiles and surface temperature from the AIRS level 2 product at the time of the satellite overpass. For the AIRS case the uncertainty in the retrieved atmospheric profiles was provided in the level 2 products.

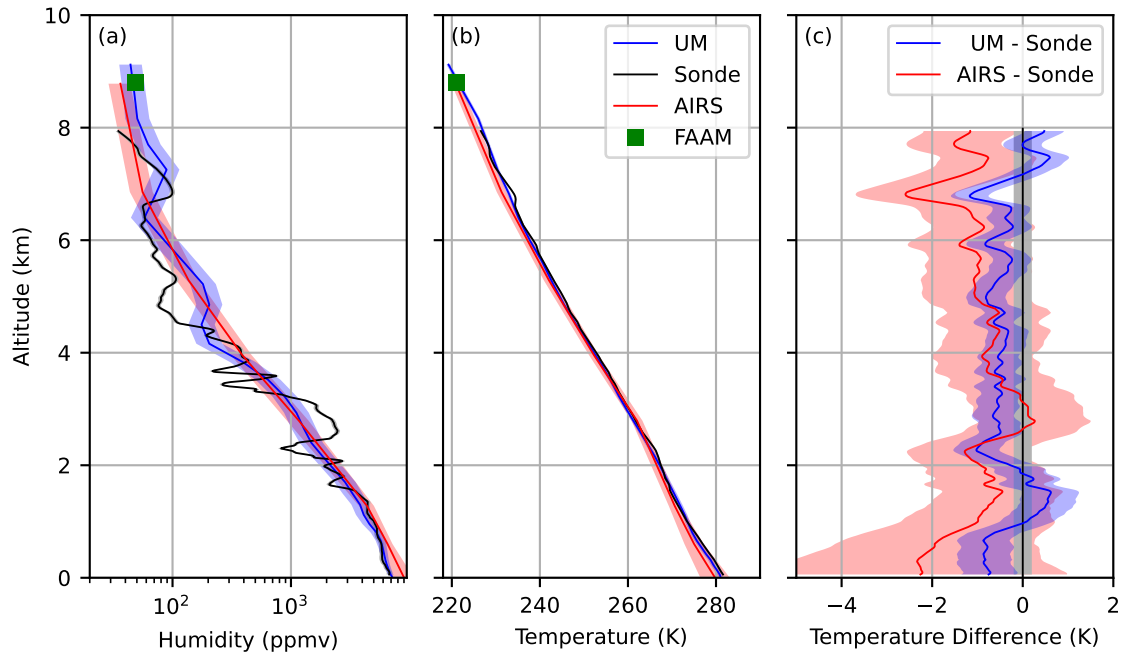


Figure 4.1: (a) Water vapour and (b) temperature profiles used as inputs for LBLRTM. In each case, the blue line is the UM forecast with blue shading representing the uncertainty from the B-matrix, the black line is the sonde with grey shading representing the uncertainty in the measurement, the red line and shading is the AIRS profile and uncertainty, and the green square is the in-situ measurements made at the aircraft. (c) Differences between the sonde temperature profile and the UM and AIRS profiles.

Figure 4.1 shows the temperature and humidity profiles used for the UM, sonde and AIRS cases.

The temperature profiles for the three cases tend to agree within the assumed uncertainties with the UM and AIRS cases generally cooler than the sonde. This may be due to the fact that the UM and AIRS profiles are average profiles over larger areas, as opposed to the point measurement made by the sonde. At around 6.8 km the sonde measures a warm region that is

not represented in the UM or AIRS cases. It is possible that this warm layer is localised (see section 4.2.6 for a discussion on the atmospheric variability). The humidity profiles for the UM and AIRS cases generally agree within the assumed uncertainty. The sonde case shows more structure with a drier layer between 4.5 and 6 km and a moist layer between 2.3 and 3.5 km.

For all cases the ozone profile was determined from the measurements made by the FAAM Airborne Laboratory TECO 49 instrument during the spiral descent later in the flight. The CO₂ was also assumed to have a constant concentration below the aircraft as described by the standard mid-latitude winter profile from Anderson et al. (1986). This concentration was chosen to be 410.6 ppm, which was the atmospheric concentration of CO₂ reported by NOAA/ESRL on the day of the flight (Dlugokencky et al., 2019). It should be noted that a test simulation of the sonde case performed using the nearest CO₂ profile in space and time from the Copernicus Atmosphere Monitoring Service global greenhouse gas reanalysis (Inness et al., 2019) produced no noticeable difference to the simulated radiance spectrum compared to the radiance spectrum produced using the fixed CO₂ concentration.

Forward modelling of the expected radiances at the aircraft level was carried out using LBLRTM v12.8 (Clough et al., 2005). Nadir radiances were generated at a resolution of 0.01 cm⁻¹ for all cases. The effect of the uncertainty in the temperature and humidity profiles on the simulated radiances was also estimated. This estimation was done by performing two additional simulations for each case with the temperature and humidity profiles altered in line with the uncertainties described above. One case was constructed to give an upper bound on the simulated spectra by increasing the temperature and decreasing the humidity and the lower bound on the simulated spectra was found by increasing the humidity and reducing the temperature. It should be noted that these radiance bounds represent a worst case scenario as it is unlikely that the error in the profile would all act in one direction. All the simulated radiances were

then Fourier transformed and apodised with both the TAFTS and ARIES ILS, producing two versions of each simulation, one to compare to TAFTS measurements and one to compare to ARIES measurements.

These simulations are compared to co-incident observations from TAFTS and ARIES taken just after the release of sonde 8. The TAFTS spectrum recorded closest to the release of sonde 8 at 12:51 UTC is used in the FIR (80 - 300 cm^{-1} and 330 - 550 cm^{-1}) and 10 averaged ARIES spectra are used in the MIR (600 - 1600 cm^{-1}). This averaging is to make the instrument noise and acquisition time of the observations more comparable, as the TAFTS spectra take longer to acquire but have a lower random error. The ARIES measurements are cut off at 1600 cm^{-1} to match the FORUM wavenumber range as this work was undertaken in support of FORUM.

4.1.1 Comparison of Simulations

The aim of this section is to compare the LBLRTM simulations of the three cases to the measured spectra and investigate whether radiance differences related to differences in the input profiles can be detected above the level of uncertainty. Figure 4.2 shows the simulated radiance for the UM, sonde and AIRS cases and the observed spectra. The figure also shows the uncertainty on the measured spectra due to the instrument noise and calibration uncertainty and the uncertainty in the simulated radiance spectra due to uncertainty in the input profiles. The observed and simulated spectra show differences in radiance in the FIR micro-windows, between 200 and 550 cm^{-1} (e.g. 240, 270, 395, 410, 520 cm^{-1}). Figure 4.3 shows a zoomed view of three of these micro-windows.

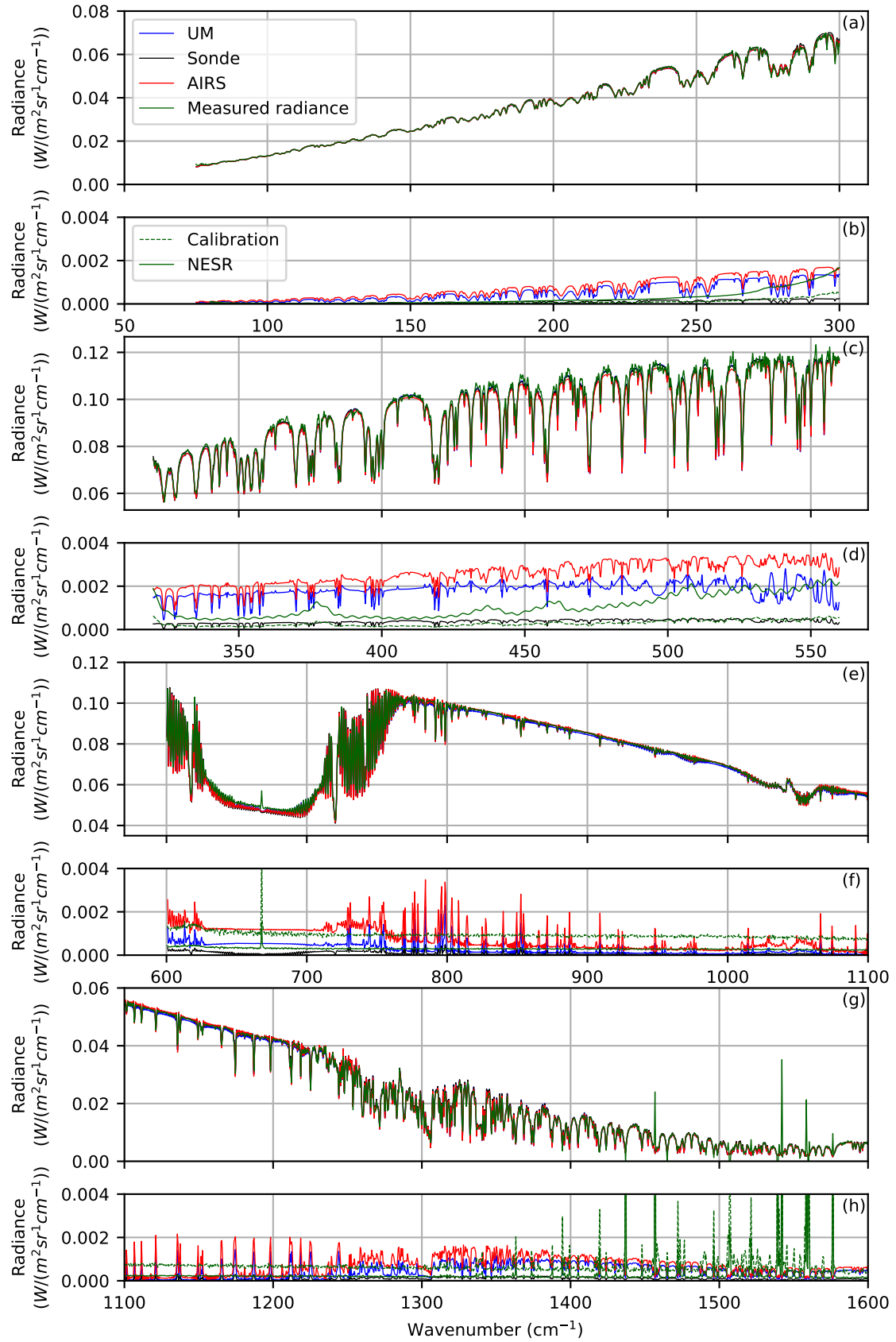


Figure 4.2: Simulated and observed radiance measurements taken at around 12:51 UTC, the time of sonde 8 release. Panels (a, c, e, g) show the spectra, and panels (b, d, f, h) show the uncertainty. Observations in the top four panels are from TAFTS, and in the bottom four panels from ARIES.

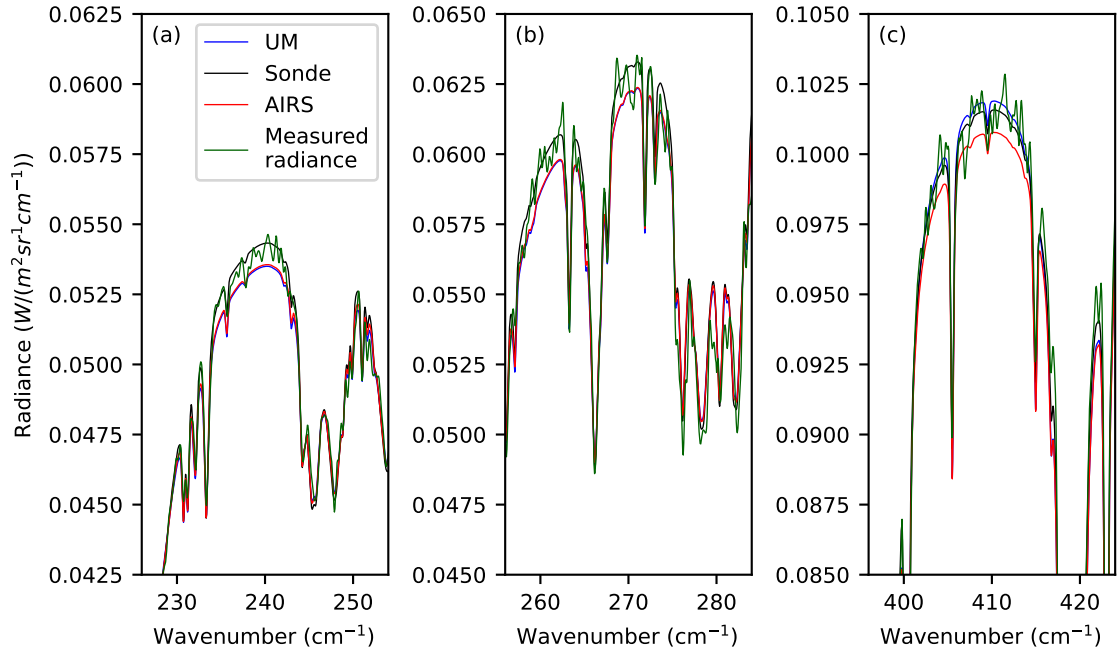


Figure 4.3: Zoomed examples of the microwindows at (a) 240, (b) 270 and (c) 410 cm^{-1} . The lines have the same colours and meaning as figure 4.2

There are also slight differences noticeable across the atmospheric window, corresponding to the different surface temperatures used in the models. In the TAFTS longwave and shortwave channels (figure 4.2b and d) the radiance uncertainty due to the uncertainty in the AIRS and UM input profiles is larger than the TAFTS NESR and calibration uncertainty. The TAFTS NESR is larger than the TAFTS calibration uncertainty and increases markedly above 450 cm^{-1} as has been seen in previous work carried out using TAFTS (e.g. Beeby, 2011; Cox et al., 2007; Fox, 2015). There are peaks in the TAFTS NESR at 360 and 440 cm^{-1} due to absorption of the Mylar beamsplitter and at 460 cm^{-1} due to absorption from the polypropylene view window. The effect of the increased measurement noise can also be seen in the observed spectrum (figure 4.2c), causing its jagged appearance above 450 cm^{-1} . The radiance uncertainty due to the uncertainty in the sonde input profile is a similar size to the TAFTS calibration uncertainty. For the ARIES measurements (figure 4.2f and h) the calibration error is larger than the NESR. In the atmospheric window this calibration error is larger than the radiance

error caused by uncertainty in the UM and AIRS input profiles. Above 1200 cm^{-1} uncertainty due to uncertainty in the input profiles becomes larger than the ARIES calibration error. Large spikes are seen in the ARIES calibration error and NESR above 1450 cm^{-1} .

Figure 4.4 shows the differences between the observed and simulated spectra with the UM case and sonde case offset by 0.02 and $0.01\text{ W m}^{-2}\text{ sr}^{-1}\text{ cm}$ respectively. The y-scales vary slightly between panels. The shading around each dashed line represents the measurement error from the instrument added in quadrature with the simulated radiance uncertainty caused by the uncertainty in the input profiles. Therefore in regions where the solid line is outside the shading, there is a difference between the measured and simulated radiance that is larger than the uncertainty. The uncertainty for the UM and AIRS cases are larger than that of the sonde case, reflecting that fact that the uncertainty on the sonde temperature and humidity profiles is smaller than that on the UM and AIRS profiles.

Between 80 and 400 cm^{-1} the three simulations tend to agree with the observations within the uncertainty. Differences between the three simulations in this region, although smaller than the total uncertainty, again are most marked within micro-windows, between 220 and 550 cm^{-1} (e.g., 230 cm^{-1} , 260 cm^{-1} , 395 cm^{-1}). In most micro-windows the sonde simulation is a better match to the measured radiances with the UM and AIRS simulations having a reduced radiance.

These differences in radiance can be linked to the differences in atmospheric profiles by considering the weighting functions, see section 1.3. Figure 4.5 shows weighting functions calculated as a function of height. The weighting functions for wavenumbers 135 and 230 cm^{-1} (figure 4.5a) peak at around 7 km but exhibit sensitivity down to an altitude of 4 km . This suggests that the radiance differences seen in these micro-windows are due to differences in the atmospheric profiles between 4 and 7 km . The higher radiance seen in the sonde simulation is consistent

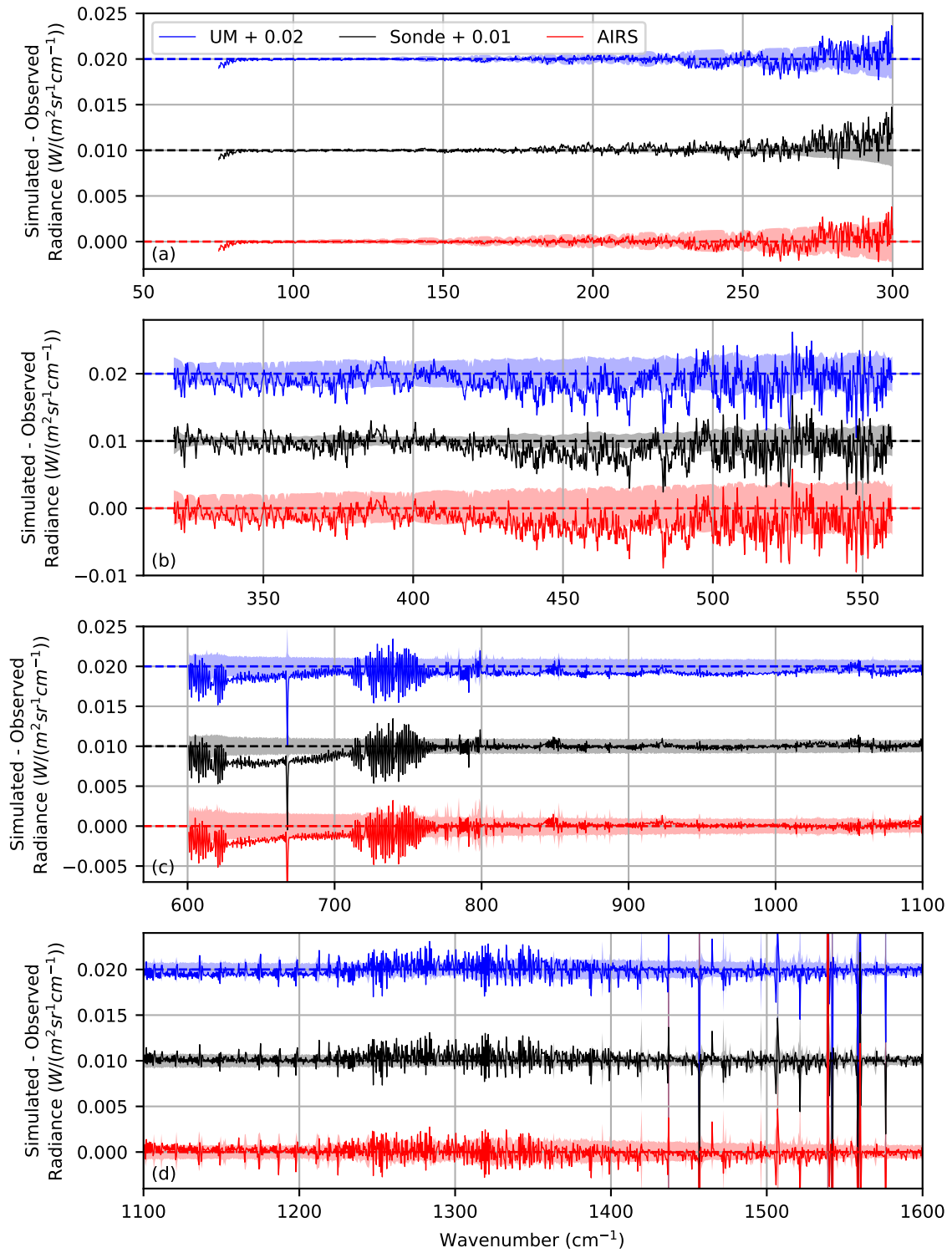


Figure 4.4: Radiance differences between UM (blue), sonde (black), AIRS (red) and TAFTS/ARIES observations (simulation - observation). The shading around the zero line represent the TAFTS/ARIES measurement uncertainty and the radiance uncertainty propagated from the uncertainty in input profile.

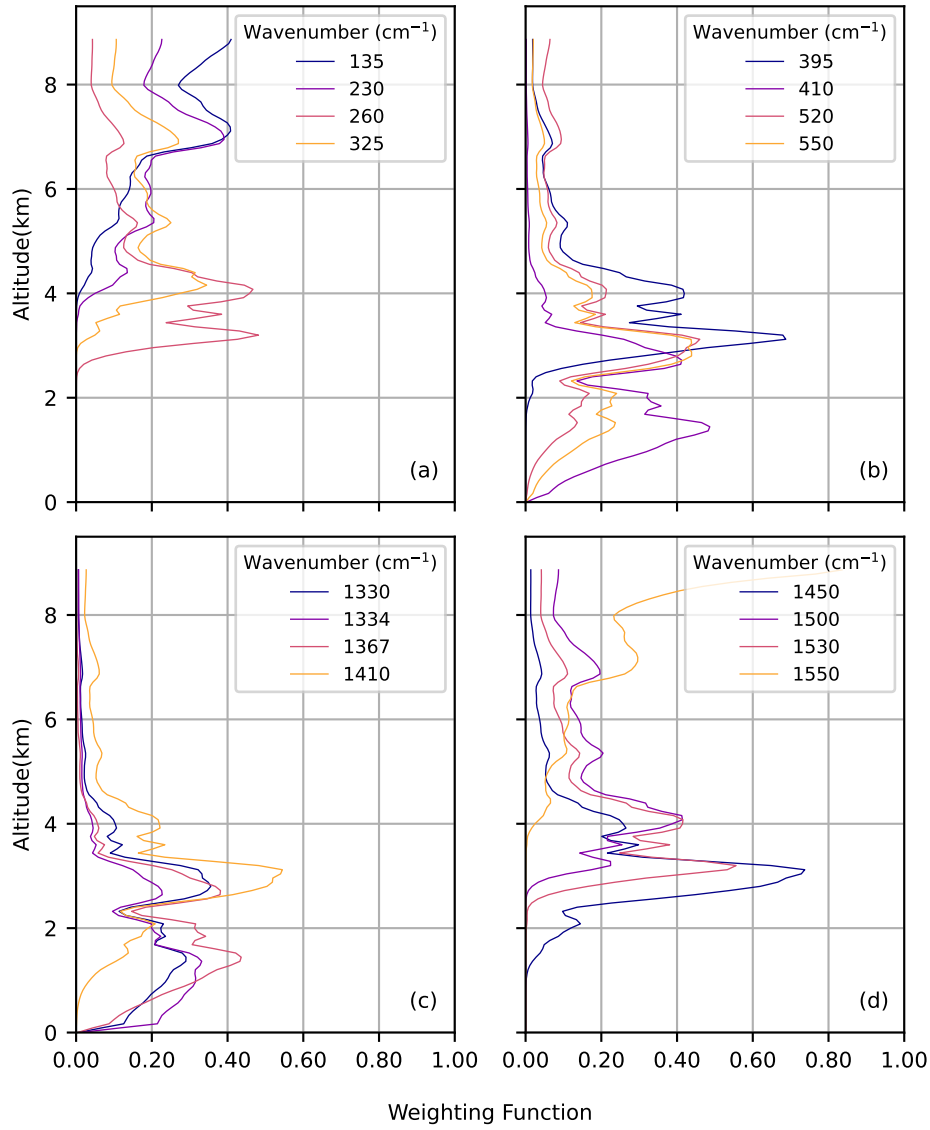


Figure 4.5: Weighting functions for wavenumbers in the (a,b) TAFTS and (c,d) ARIES wavenumber ranges.

with the drier profile seen in the sonde case. As the wavenumbers increase, the weighting functions show that the radiance is sensitive to the profile lower in the atmosphere. For example the weighting function corresponding to the 395 cm^{-1} micro-window peaks at around 3.5 km (figure 4.5b).

Returning to the radiance difference plot (figure 4.4b), between 320 and 400 cm^{-1} the UM and AIRS simulations tend to show a negative bias that is less marked in the sonde case. Looking at the weighting functions for 325 and 395 cm^{-1} this suggests the radiance simulations are

sensitive to the differences in the input profiles between 3 and 7 km and that in this altitude range the sonde profile is the best match for the atmosphere observed by TAFTS. Between 425 and 550 cm^{-1} all simulated cases exhibit a negative bias of 3-5 $\text{mW m}^{-2} \text{sr}^{-1} \text{cm}$ but show relatively good agreement with each other. The weighting functions suggest this region of the spectrum is sensitive to the atmospheric profile at around 3 km and below. However, a similar underestimation is not noticeable in the region of the ARIES spectrum that sounds this region of the atmosphere (1300-1400 cm^{-1} , figure 4.5c). Cox et al. (2007) also observed differences between clear sky simulations and TAFTS spectra in this wavenumber region, however the differences were in the strong water vapour lines as opposed to the more broadband difference seen here.

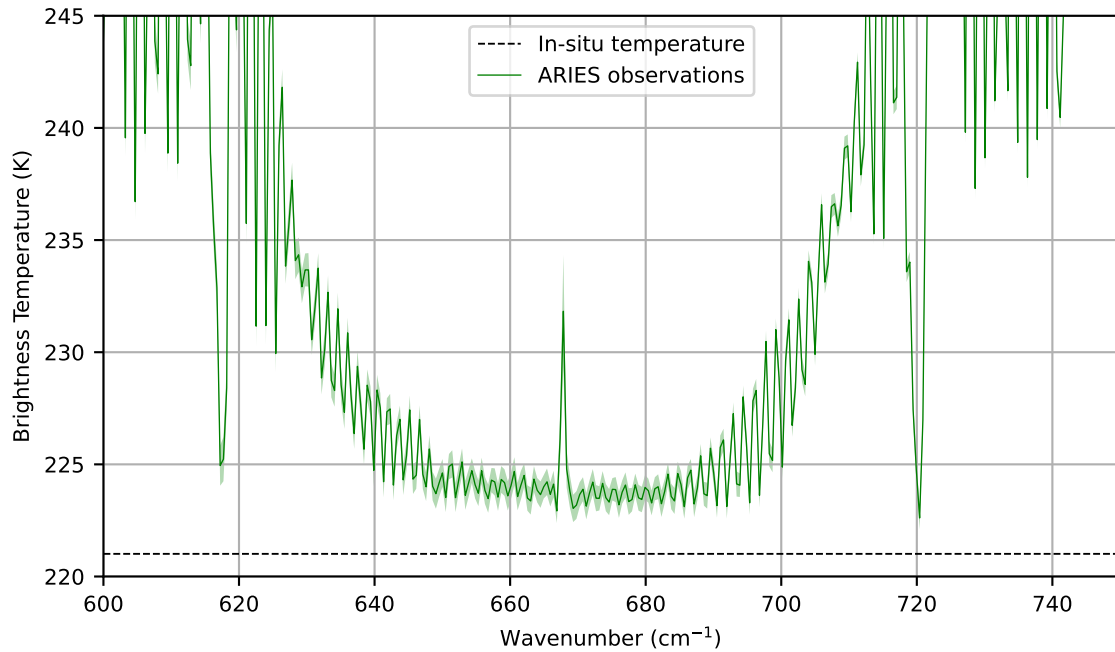


Figure 4.6: Detail of the ARIES spectrum around the 667 cm^{-1} CO_2 band in units of brightness temperature. The dashed line shows the temperature measurement from the in-situ sensor on the FAAM aircraft.

Turning to the MIR (figure 4.4c and d), the largest difference between the simulations and observations occurs in the CO_2 band from 650-700 cm^{-1} , a region which is sensitive to the properties of the atmosphere very close to the aircraft. The radiance simulated in all three cases

is markedly lower than that observed. Analysis of the corresponding brightness temperature, figure 4.6, shows that the ARIES observations are around 3 K warmer than the outside air temperature measured by the in-situ aircraft sensors. It is likely that heating of the instrument housing, as has been observed in previous measurements (Wilson et al., 1999), is causing this effect.

A difference of approximately $1 \text{ mW m}^{-2} \text{ sr}^{-1} \text{ cm}$ between the UM simulation and the other simulations is seen across the atmospheric window between 800 and 1200 cm^{-1} . This difference is caused by the cooler surface temperature predicted by the UM forecast. Progressing into the wing of the vibration-rotation band of water vapour ($1200\text{-}1350 \text{ cm}^{-1}$), all three simulations show a slight positive bias compared to the observations with some features extending above the uncertainty. The weighting functions for this wavenumber range are plotted in figure 4.5c and show that this spectral range is sensitive to the atmospheric profile between 1 and 4 km. Above 1400 cm^{-1} the UM and AIRS simulations show a slight negative bias in comparison to the sonde simulation. The weighting functions in figure 4.5d show that this is linked to the atmospheric profile between 3 and 6 km. This is therefore consistent with the negative bias seen in the FIR observations between 320 and 400 cm^{-1} , however the difference is smaller in the MIR than the FIR and also small compared to the uncertainty.

4.1.2 Sensitivity Studies

Overall, the similarity in residual shape for all three cases between 425 and 550 cm^{-1} , without a similar underestimation in the MIR, indicates that there is either some systematic error affecting all the simulations or an underestimate of the instrument uncertainty. The size of this bias is $3\text{--}5 \text{ mW m}^{-2} \text{ sr}^{-1} \text{ cm}$. To understand this difference four further forward modelling

studies were undertaken investigating the impact of changing the surface temperature, surface emissivity, water vapour continuum and water vapour line widths.

Sensitivity of Spectra to Surface Temperature and Emissivity

The aim of this experiment was to investigate the sensitivity of simulated spectra to changes in surface temperature and emissivity. Surface conditions are primarily retrieved using the measured radiance in the atmospheric window where transmission through the atmosphere is high meaning that the signal from the surface is strong (section 1.3). However, some radiance from the surface can be detected in the FIR micro-windows (e.g. 495 cm^{-1} , 530 cm^{-1} , 560 cm^{-1}) particularly in dry conditions (Feldman et al., 2014). Therefore, uncertainty in the surface conditions could impact the simulated spectra in the FIR and may explain the differences between simulations and measurements between 400 and 550 cm^{-1} .

First, the effect of surface temperature was investigated. This was done by producing three additional simulations using the atmospheric profiles from the sonde case, with only surface temperature altered. Three surface temperatures were used (280.50 , 280.75 , 281.00K) chosen so they would produce radiance signals within the ARIES measurement uncertainty across the atmospheric window. Figure 4.7 shows the differences between these simulations and the simulation using the original surface temperature of 280.25 K . As expected, the differences are most noticeable in the atmospheric window, however some change in radiance is noticeable in the FIR between 400 and 600 cm^{-1} , reaching a maximum value of $0.4\text{ mW m}^{-2}\text{ sr}^{-1}\text{ cm}$ at 550 cm^{-1} . However, this radiance difference caused by changing surface temperature is at most 10% of the bias seen between 425 and 550 cm^{-1} . Therefore, a mischaracterisation of surface temperature could only be contributing a very small amount to the bias seen in this region.

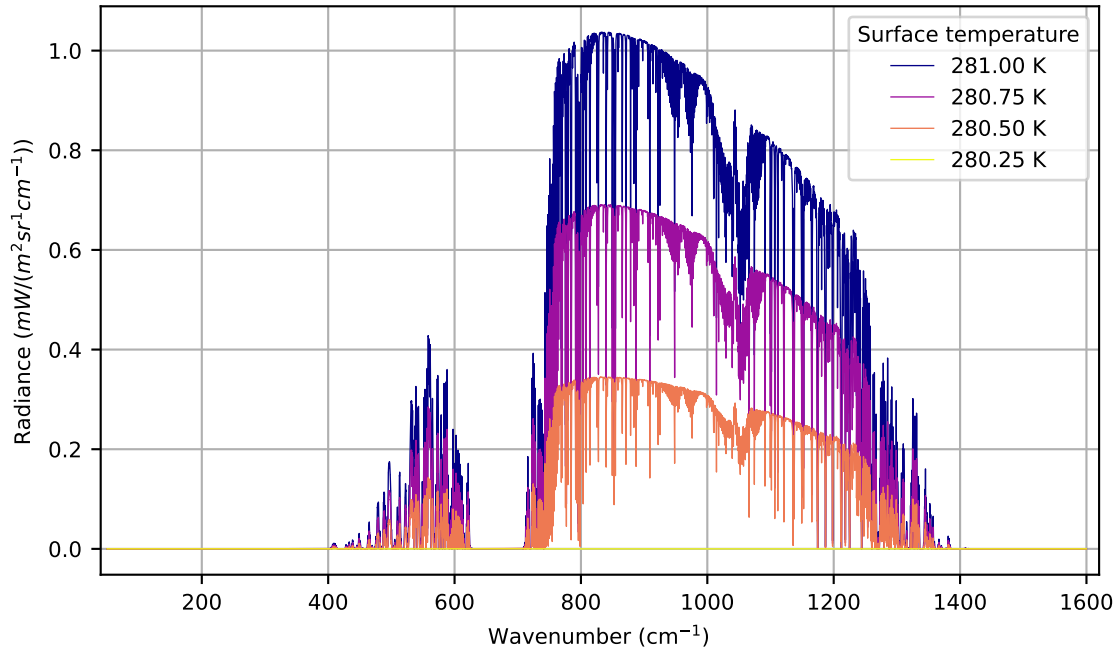


Figure 4.7: Difference in radiance from the sonde simulation caused by changes in the surface temperature.

However, as the measurements between 425 and 550 cm^{-1} were sensitive to the surface conditions a second test was carried out to investigate the effect of surface emissivity on the expected FIR radiance. As the emissivity of the ocean in the FIR has not been experimentally determined (see chapter 6) uncertainty in the surface emissivity used in the simulation could be a cause for some of the apparent bias in the TAFTS observations. To test this a simulation was performed using the atmosphere and surface temperature of the sonde case but with an emissivity of 1 throughout the spectrum, representing the absolute upper bound of the possible emissivity. Figure 4.8 shows the radiance difference between these simulations and the simulations with the IREMIS emissivity. The change in emissivity causes a change in radiance of at most $1.5 \text{ mW m}^{-2} \text{ sr}^{-1} \text{ cm}$ at 550 cm^{-1} . This represents around 40% of the bias seen in this region, however this is likely an overestimate of the contribution as the theoretical calculations such as those from the Huang dataset suggest that the emissivity of pure water is less than one in this region (Huang et al., 2016).

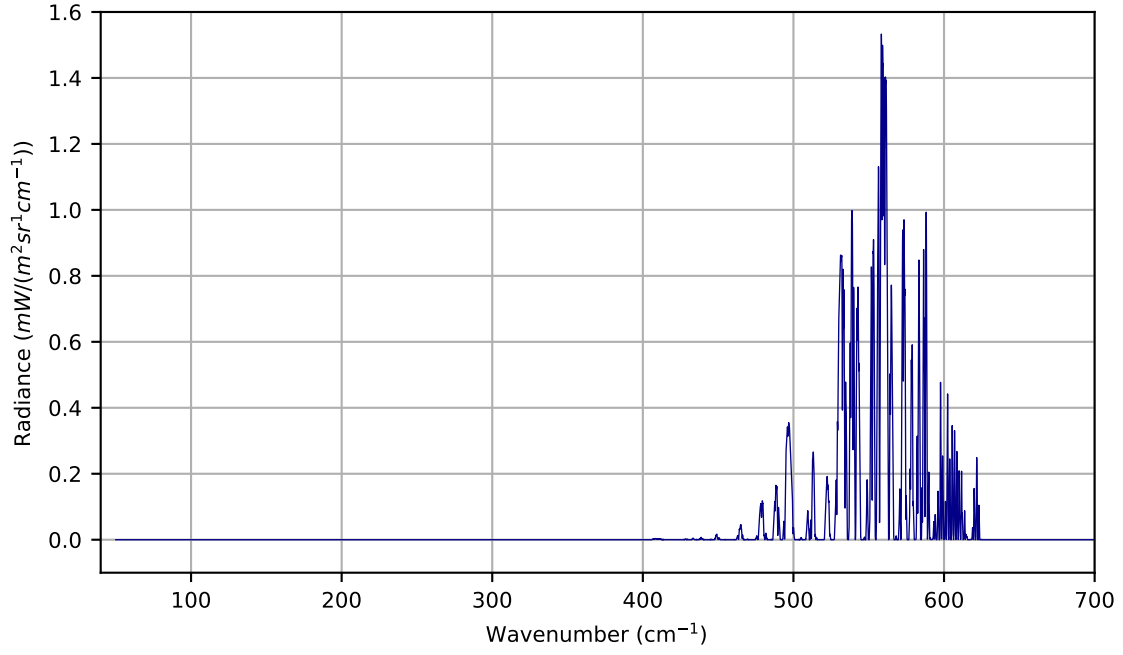


Figure 4.8: Difference in radiance from the sonde simulation caused by assuming an emissivity of one.

Overall, in a worst case scenario, uncertainty in the surface conditions could explain up to 50% of the bias seen in the TAFTS spectrum between 425 and 550 cm^{-1} .

Sensitivity of Spectra to Spectroscopy

Another phenomenon which could be causing systematic changes to the simulations, is a misrepresentation of the water vapour continuum (Harries et al., 2008). There is still an uncertainty of up to 20% in the strength of the water vapour continuum at wavenumbers below 400 cm^{-1} and of at least 7% at wavenumbers between 400 and 600 cm^{-1} (Mlawer et al., 2019), which could cause differences between simulations and observations. For this study, the strength of the water vapour continuum in LBLRTM was adjusted by $\pm 10\%$ and $\pm 20\%$ while all other aspects of the simulation were kept the same as in the sonde case. Figure 4.9 shows the impact of these changes on simulated spectra.

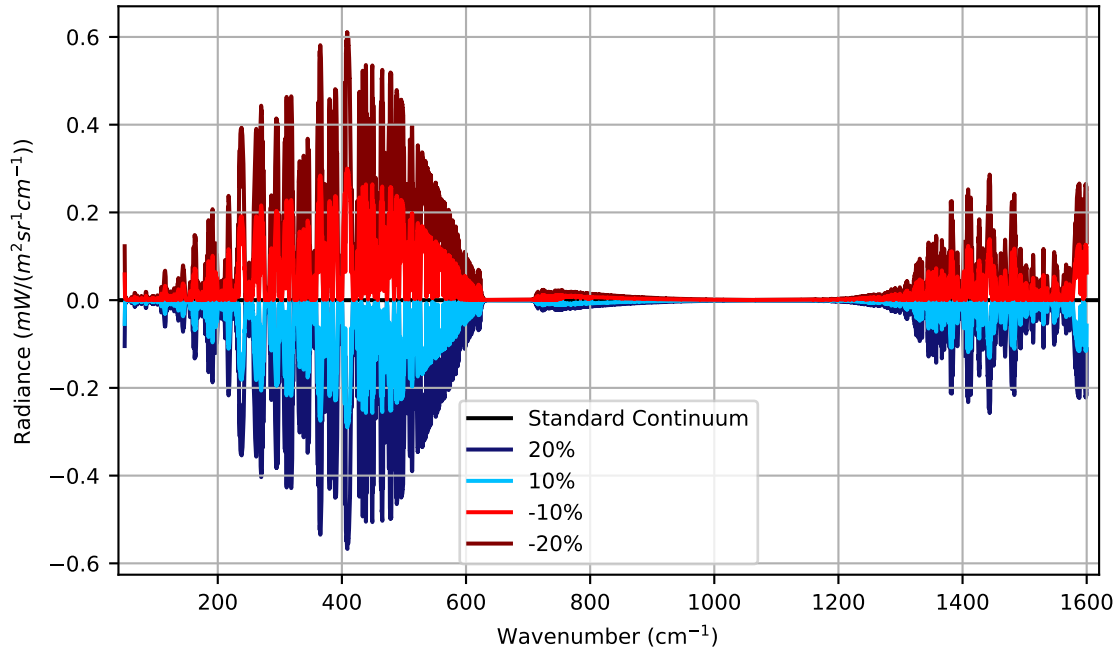


Figure 4.9: Change in radiance from the sonde simulation caused by altering the strength of the water vapour continuum.

The uncertainty in the continuum has the largest impact between 300 and 500 cm^{-1} with significant changes noticeable throughout the FIR and some slight differences noticeable in the vibration-rotation water vapour band between 1300 and 1600 cm^{-1} . Between 425 and 500 cm^{-1} an uncertainty of 10% in the water vapour continuum translates to a change of at most $0.25 \text{ mW m}^{-2} \text{ sr}^{-1} \text{ cm}$ in the observed spectrum, roughly 6% of the observed bias.

As well as uncertainty in the strength of the water vapour continuum, there is also uncertainty in the water vapour line widths that are reported in the AERv3.8 database (the H_2O line parameters in AERv3.8 are based on the HITRAN 2012 database which is used for LBLRTM (Rothman et al., 2013)). Alessio Di Roma performed three simulations to estimate the impact of this uncertainty: an unperturbed case, a case where all the water vapour line widths were reduced by the quoted uncertainty and a case where all water vapour line widths were increased by the quoted uncertainty. The atmospheric profiles and surface properties were the same as those

used in the sonde simulation. Figure 4.10 shows the change in radiance caused by this change in spectroscopic parameters.

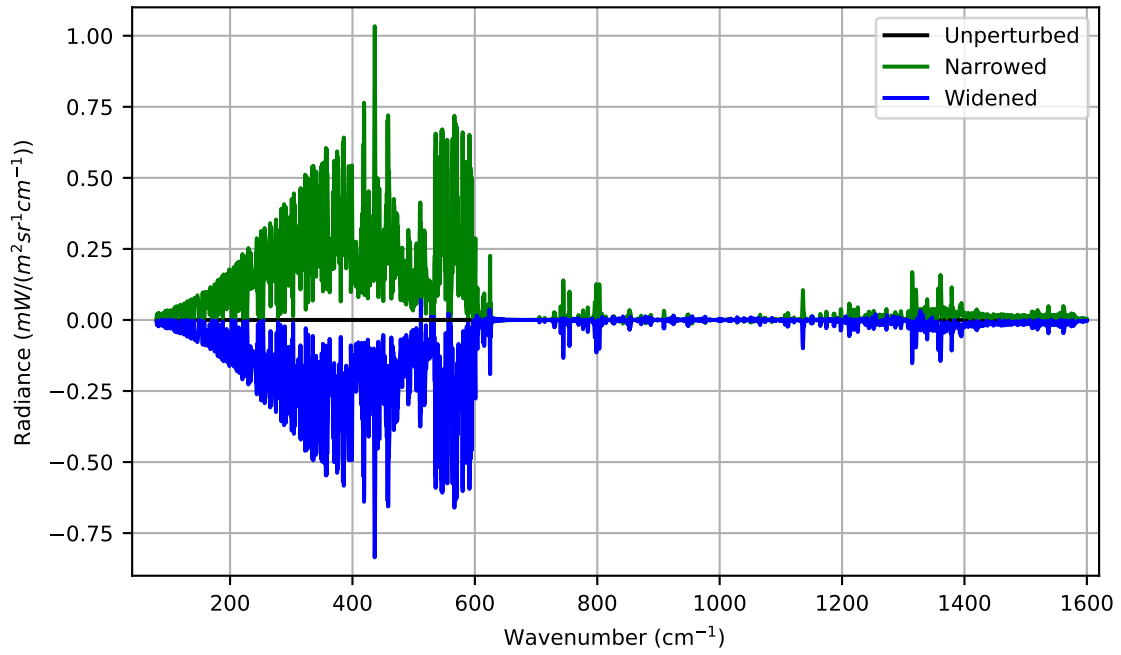


Figure 4.10: Change in radiance from the sonde simulation caused by altering the widths of the water vapour lines.

The maximum value of the radiance change caused by a change in water vapour line widths is $0.7 \text{ mW m}^{-2} \text{ sr}^{-1} \text{ cm}$ at around 430 cm^{-1} but it is mostly below $0.5 \text{ mW m}^{-2} \text{ sr}^{-1} \text{ cm}$ between 425 and 500 cm^{-1} . This is around 12% of the observed bias. Together this implies that spectroscopic effects could account for around 18% of the difference seen between the simulations and measurements.

Conclusion of Sensitivity Studies

The overall maximum difference in radiance found by summing all the perturbations considered in this section is around $2.7 \text{ mW m}^{-2} \text{ sr}^{-1} \text{ cm}$. This is just smaller than the minimum size of the bias between the observations and simulations which was between 3 and $5 \text{ mW m}^{-2} \text{ sr}^{-1} \text{ cm}$.

Therefore it is unlikely that uncertainty in the surface characteristics or spectroscopy can fully explain the bias seen between 425 and 550 cm^{-1} . Unless the uncertainties in the water vapour line parameters and spectroscopy are larger than reported, which is possible, it would appear that the uncertainties estimated for the TAFTS measurements in this region are too small. As this difference affects higher wavenumbers this may be due to the effect of aircraft vibrations causing “ghosts” that were not removed by the “de-ghosting” procedure mentioned in section 3.1.1. It should also be noted that the uncertainties associated with the sonde temperatures and humidities do not consider the impact of drift of the sonde as it falls. See section 4.2.6 for a discussion of this sonde drift.

4.1.3 Conclusions of Forward Modelling

Overall, the simulations in this section have demonstrated that the FIR is sensitive to differences between the temperature and water vapour profiles in the three cases tested. A possible underestimation of calibration errors in the TAFTS spectrum between 425 and 500 cm^{-1} has been identified, which cannot be entirely explained by uncertainty in surface conditions or spectroscopy. The measured ARIES radiance in the 667 cm^{-1} CO_2 band is also higher than expected given the in-situ temperature measurements, likely caused by local heating of the instrument housing.

4.2 Retrievals

Alongside the forward modelling, colleagues from the Istituto di Scienze dell’Atmosfera e del Clima (ISAC-CNR) Bologna, Italy and the Istituto per le Applicazioni del Calcolo (IAC-CNR)

Sesto Fiorentino, Italy performed retrievals of the temperature and water vapour profiles from the measured radiances. I selected and prepared the input data for the retrievals. These data were then sent to our colleagues who performed the retrievals and sent back the output files. I analysed the output of the retrievals including calculating the DFS and, in discussion with our colleagues, designed some additional tests (see sections 4.2.4 and 4.2.5) to further investigate the retrievals.

4.2.1 Retrieval Method

The Level 2 Module – Inversion (L2M_I) code, written for the FORUM End-to-End simulator project, was used to perform the retrievals. The purpose of the End-to-End simulator is to simulate and evaluate the performance of the FORUM L1 and L2 products (Sgheri et al., 2022). It therefore contains the inversion module L2M_I to perform retrievals using the simulated instrument measurements. The L2M_I module can perform retrievals of surface parameters, such as surface temperature and emissivity, and vertical profiles, such as temperature and water vapour mixing ratio, in clear or cloudy conditions. The code can also take co-located observations from up to five sensors as inputs.

The retrieval code is based on the optimal estimation method described by Rodgers (2000) and outlined in section 2.2. LBLRTM v12.8 is used as the Forward Model of the L2M_I to produce simulated radiance spectra and Jacobians at a resolution of 0.001 cm^{-1} . These simulated spectra are convolved with the provided ILS(s) to obtain simulated spectra and Jacobians at the resolution of the instrument(s). The simulated spectra are then compared to the measured spectra and the difference is minimised using the Gauss-Newton algorithm with Levenberg optimization, see equation 2.28 (Sgheri et al., 2022).

4.2.2 Evaluation Metrics

In this chapter, two metrics will be used to assess the performance of the retrievals: the degrees of freedom of signal (DFS) and the root mean squared error (RMSE).

The DFS is a measure of the amount of information added by a particular observation. It incorporates the variance-covariance matrices of the measurement and the a-priori, and the Jacobian matrix so it depends on both the instrument characteristics and how inherently sensitive the radiance is to changes in the atmospheric profile (section 2.2).

The RMSE is calculated from the difference between the profile of interest and the sonde profile, which is assumed to represent the true atmospheric state (see section 4.2.6 for a discussion of the role of atmospheric variability). The RMSE values for water vapour are calculated using the units of the logarithm of specific humidity, as the water vapour concentration varies by several orders of magnitude over the height of the profile. This is appropriate as the logarithm of specific humidity is a good measure of water vapour radiative impact (Soden & Bretherton, 1993). To assess the vertical performance of the retrieval, the calculation is split into different height regions. It should be noted that conclusions about the performance of the retrieval inferred from the RMSE values are likely specific to the atmospheric conditions sampled in this study.

4.2.3 Temperature and Water Vapour Retrievals

Using the L2M.I code, joint retrievals of atmospheric temperature, water vapour and surface skin temperature were performed. Two retrievals were performed, one using the TAFTS spectrum and one using the average of the ten ARIES spectra as described in section 4.1. In

both cases the a-priori temperature and water vapour profiles and surface skin temperature were taken from the Met Office UM as shown in figure 4.1. The spectral variation in surface emissivity was derived as described in section 4.1 and was fixed in the retrieval. The a-priori variance-covariance matrix was adapted from the UM B-matrix. When these retrievals were performed the L2M_I code required that the variance-covariance matrix be defined by specifying the a-priori error for each retrieved parameter and a correlation length, rather than providing the matrix directly. It was found that a correlation length of 0.8 km best reproduced the UM B-matrix. This method for specifying the variance-covariance matrix meant that cross correlation between temperature and water vapour could not be specified in the retrieval and was therefore assumed to be zero. This is justified because the magnitude of the cross correlation was many orders of magnitude smaller than the magnitude of the correlation within the temperature and water vapour profiles. The measurement variance-covariance matrices for the TAFTS and ARIES retrievals were constructed from the total measurement error of the TAFTS and ARIES instruments and were assumed to be diagonal.

The water vapour and temperature profiles for the TAFTS retrieval are shown in figure 4.11. The a-priori and sonde profiles are identical to those in figure 4.1. The relatively dry layer measured by the sonde between 4 and 6 km is captured well by the TAFTS humidity retrieval with the retrieved profile moving towards the sonde measurement and away from the a-priori. However, the retrieval cannot capture the moist layer at 3 km and instead produces a moist layer below this that is not present in the sonde measurements. This is accompanied by a positive bias in the temperature retrieval which reaches over 2 K between 0 and 1 km. Conversely, between 3 and 7 km the retrieved temperature profile is cooler than that measured by the sonde. Looking at the RMSE values for the a-priori and retrieved profiles, table 4.2, confirms this visual impression.

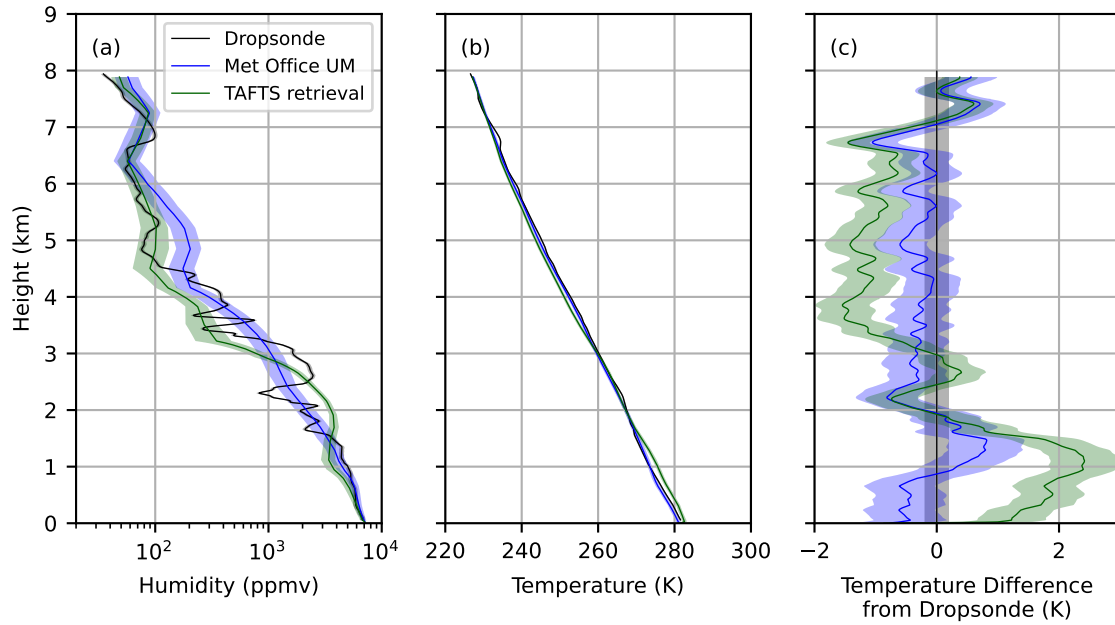


Figure 4.11: (a) Water vapour and (b) temperature retrievals (green) performed using the TAFTS radiances. The green shading around the line represents the uncertainty in the retrieval taken from the diagonal elements of the a-posteriori error covariance matrix. The blue line is the a-priori profile and the blue shading the uncertainty in the a-priori profile. The black line is the profile measured by sonde 8 with the grey shading representing the limits of the measurement uncertainty, which is significantly smaller than the a-priori or retrieval uncertainty. Panel (c) shows the differences of the a-priori and retrieved temperature profiles from the sonde 8 temperature profile.

RMSE	Water Vapour ($\log(\text{kg/kg})$)			Temperature (K)		
	UM	Full range	Reduced range	UM	Full range	Reduced range
Overall	0.42	0.40	0.34	0.44	1.23	0.51
0 – 2 km	0.11	0.28	0.19	0.50	1.72	0.49
2 – 4 km	0.50	0.61	0.49	0.42	0.84	0.60
4 – 6 km	0.63	0.34	0.29	0.33	1.15	0.38
6 – 8 km	0.31	0.24	0.34	0.46	0.70	0.52

Table 4.2: RMSE values for the UM, TAFTS full range and TAFTS reduced range retrievals. All RMSE values are calculated compared to the sonde 8 profiles.

Generally, the TAFTS water vapour retrieval has a lower RMSE than the a-priori UM profile, with the improvement mainly occurring between 4 and 8 km. Below 4 km the retrieval is further from the sonde than the a-priori. The RMSE values for the temperature retrieval are larger than those for the a-priori with the bottom 2 km of the retrieval having particularly large

RMSE values.

Considering the weighting functions in figure 4.5, this bias in the retrieval below 3 km is likely caused by the bias seen in the TAFTS measurements between 425 and 550 cm^{-1} . Therefore, the TAFTS retrieval was re-run using a reduced wavenumber range of 100-440 cm^{-1} . All other elements of the retrieval were left unchanged. The second retrieval will be called the TAFTS reduced range retrieval in the rest of this discussion.

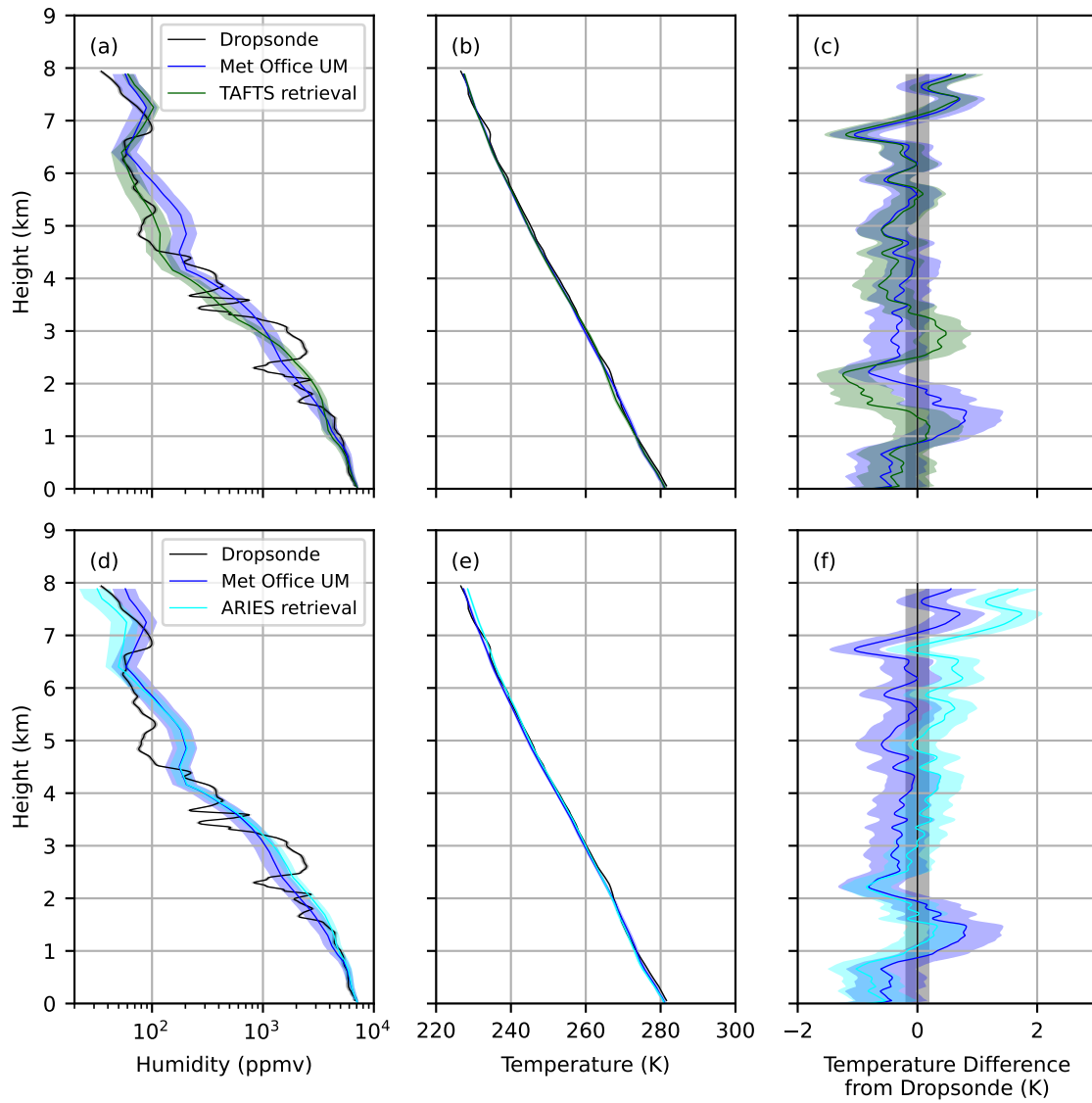


Figure 4.12: (a) Water vapour and (b) temperature retrievals performed using the reduced range TAFTS spectrum. The a-priori and sonde profiles are also shown. The shading around each line has the same meaning as figure 4.11. Panel (c) shows the differences of the a-priori and retrieved temperature profiles from sonde 8. (d-f) as (a-c) but for the ARIES retrieval.

The resulting TAFTS reduced range retrieval can be seen in the top row of figure 4.12. Removing the TAFTS wavenumbers above 440 cm^{-1} markedly reduces the temperature bias in the bottom 2 km of the atmosphere and improves the agreement with the sonde between 3 and 7 km. The water vapour retrieval also shows improved agreement with the sonde between 1 and 3 km while still following the drier sonde profile between 4 and 6 km. This removal of the TAFTS wavenumbers above 440 cm^{-1} reduces the RMSE for the temperature throughout the atmosphere, most notably between 0 and 2 km, see table 4.2. The RMSE is also reduced for the water vapour profile as a whole and at all heights below 6 km.

Comparing the ARIES (bottom row of figure 4.12) and TAFTS retrievals, the most notable difference occurs in the water vapour profile between 4-6 km, where the ARIES retrieval is much closer to the a-priori UM forecast and the TAFTS retrieval is closer to the sonde profile. A second difference is seen in the temperature profiles above 7 km, where the ARIES retrieval is significantly warmer than the sonde, a-priori and the TAFTS retrieval. This is likely caused by the higher radiance measured across the centre of the 667 cm^{-1} CO_2 band (see figure 4.6), as this region of the spectrum sounds the atmosphere close to the radiance measurements.

Comparing the RMSE values, table 4.3, shows that the RMSE values are lower for the TAFTS reduced range retrievals than the ARIES retrievals for both water vapour and temperature. This advantage is retained for the majority of the vertical layers considered.

Table 4.4 contains the DFS values for the TAFTS full range, TAFTS reduced range and ARIES retrievals (rows 1, 2 and 4). Comparing the TAFTS full range and reduced range retrievals, the reduction in spectral range does lead to a reduction in DFS across all three parameters. This reduction is largest for surface temperature showing that the TAFTS measurements below 440 cm^{-1} are not very sensitive to changes in the surface properties. This is expected given the

forward modelling in section 4.1.2. The reduction in DFS between the TAFTS full range and reduced range retrievals for the water vapour and temperature profiles is less than 0.2. This indicates that the reduced range TAFTS retrieval still contains the majority of the information about the atmospheric profile, as expected from the shapes of the retrieved profiles.

RMSE	Water Vapour ($\log(\text{kg/kg})$)			Temperature (K)		
	UM	TAFTS	ARIES	UM	TAFTS	ARIES
Overall	0.42	0.34	0.42	0.44	0.51	0.60
0 – 2 km	0.11	0.19	0.42	0.50	0.49	0.54
2 – 4 km	0.50	0.49	0.50	0.42	0.60	0.35
4 – 6 km	0.63	0.29	0.63	0.33	0.38	0.32
6 – 8 km	0.31	0.34	0.25	0.46	0.52	1.04

Table 4.3: RMSE values for the UM, TAFTS reduced range and ARIES retrievals. The UM and TAFTS reduced range values can also be seen in table 4.2.

DFS	$\text{H}_2\text{O}(z)$	$T(z)$	T_{skin}
TAFTS (Full Range)	5.282	2.812	0.946
TAFTS (Reduced Range)	5.129	2.640	0.035
TAFTS (1 cm^{-1})	4.521	2.140	0.008
ARIES	3.034	2.411	0.999
ARIES (Half Calibration)	4.117	3.414	0.999

Table 4.4: DFS values for water vapour and temperature profiles and surface skin temperature for the TAFTS full range, reduced range and 1 cm^{-1} retrievals and the ARIES and ARIES half calibration error retrievals.

As expected the DFS value for surface temperature is higher in the ARIES retrieval than either TAFTS retrieval, due to ARIES covering the atmospheric window. However, the ARIES DFS values for temperature and water vapour profiles are lower than those for the reduced range TAFTS retrieval. This temperature result is somewhat surprising given that the centre and

one wing of the 667 cm^{-1} CO_2 band is included in the ARIES wavenumber range. This should give ARIES the ability to sound the atmospheric temperature throughout the profile.

4.2.4 Additional Retrievals

Because of the unexpectedly low DFS reported for the ARIES temperature retrieval further retrievals were carried out to try and understand this behaviour.

The first hypothesis was that the higher spectral resolution of TAFTS was the cause of the increased DFS. Therefore the spectral resolution of the TAFTS radiances was reduced from 0.24 cm^{-1} to 1 cm^{-1} to match that of ARIES. The instrument noise from TAFTS was reduced to account for the spectral averaging at the lower resolution and the TAFTS calibration error was held fixed. All other input to the retrieval were the same as the previous retrievals. This configuration will be referred to as the TAFTS 1 cm^{-1} retrieval.

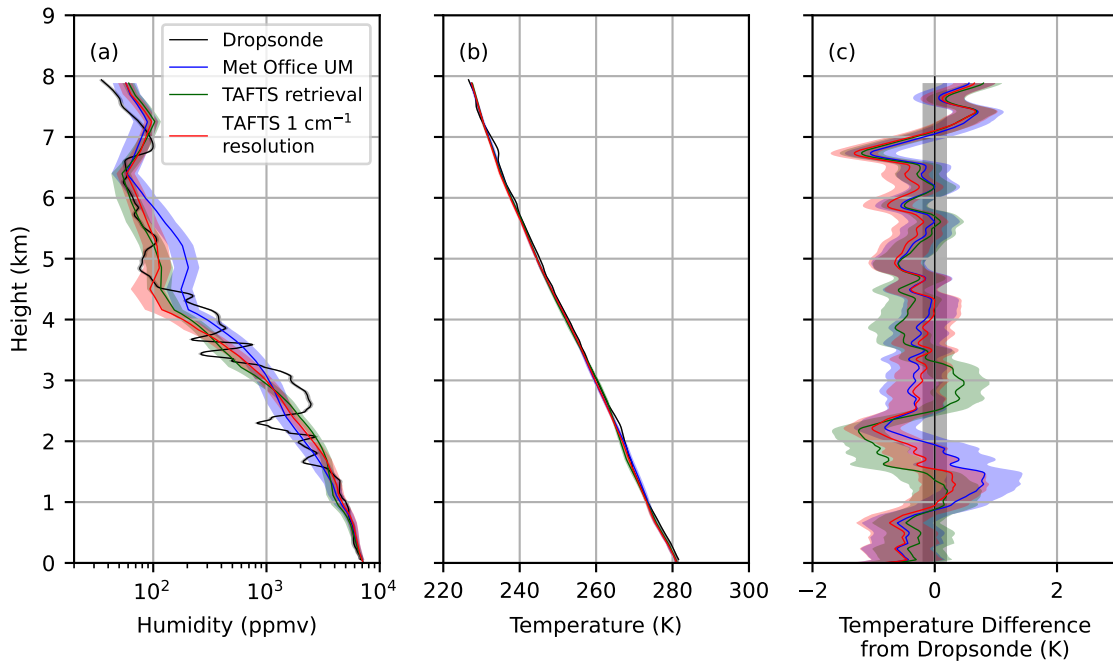


Figure 4.13: As figure 4.12 (a-c) but also showing the TAFTS 1 cm^{-1} water vapour and temperature retrievals.

Figure 4.13 shows the resulting temperature and water vapour retrievals for the TAFTS 1 cm^{-1} retrieval compared to the TAFTS reduced range retrieval. The shapes of the two retrievals are similar, suggesting that the performance of the TAFTS retrieval, particularly its ability to match the sonde water vapour measurements between 4 and 6 km, has not been significantly affected by the reduced resolution. However the DFS values, see table 4.4 row 3, are slightly reduced compared to the TAFTS reduced range case. This means that the DFS for the ARIES temperature retrieval are higher than the TAFTS 1 cm^{-1} case. However, the DFS for water vapour for the TAFTS 1 cm^{-1} case are still markedly higher than those for ARIES, indicating that at an equivalent resolution the TAFTS water vapour retrievals still contain more information about the water vapour profile.

Although the first test showed that the higher TAFTS resolution could explain the higher TAFTS DFS values for temperature, the low DFS values for the equivalent ARIES retrieval should still be explained. As the DFS values are partially dependent on the instrument uncertainty we decided to test if the ARIES uncertainty could be reduced, and thus the DFS values increased, without increasing the RMSE in the retrievals. Therefore the ARIES retrieval was rerun with the calibration error reduced by 50%.

Figure 4.14 shows this retrieval compared to the original ARIES retrieval. Although the DFS values have increased, see table 4.4 row 5, the performance of the temperature retrieval is worse, having shifted further from the sonde measurements throughout the profile.

Overall from these two additional tests it can be seen that the poor performance of the ARIES temperature retrieval compared to the TAFTS reduced range retrieval is likely caused by a combination of the higher TAFTS resolution and the higher ARIES calibration uncertainty. However when the ARIES error and the TAFTS resolution are reduced the TAFTS water vapour

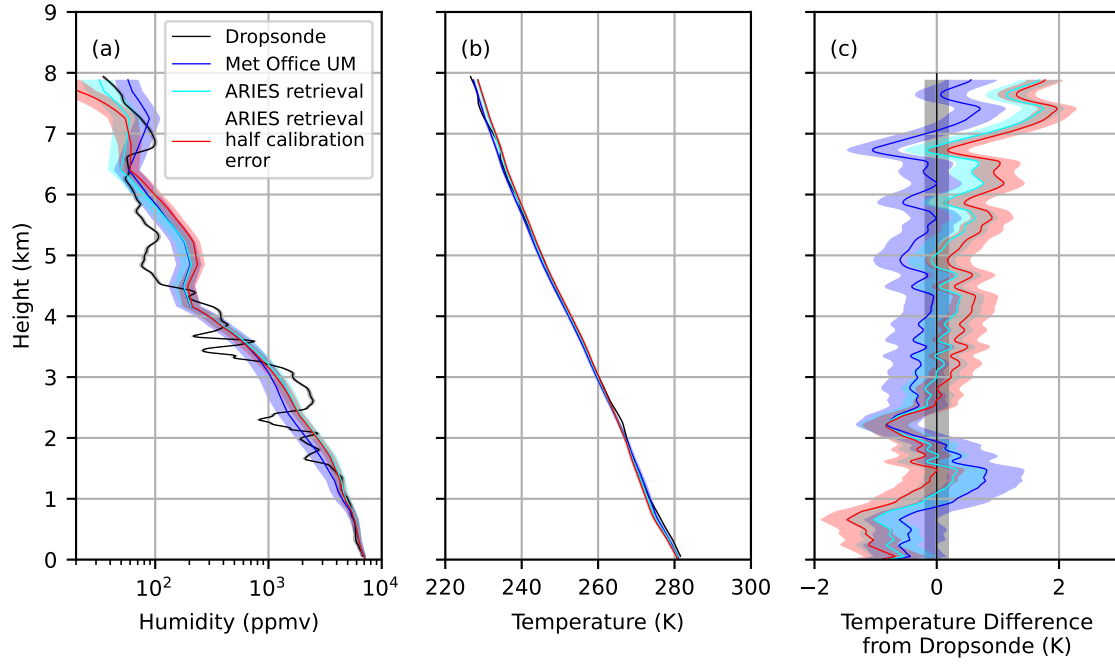


Figure 4.14: As figure 4.12 (d-f) but also showing the ARIES half calibration error water vapour and temperature retrievals.

retrieval still has a higher DFS value than the ARIES retrieval indicating that our finding of increased water vapour profile information in the FIR region of the spectrum is robust.

4.2.5 Fixed Temperature Retrievals

Due to the poor performance of the ARIES temperature retrieval caused by a combination of the high (but realistic) calibration error and offset in the 667 cm^{-1} CO_2 band we decided to perform a final set of retrievals of water vapour and surface skin temperature only. This was done by fixing the temperature profile to that measured by the sonde. Retrievals were performed using the TAFTS reduced range and ARIES spectra and a joint spectrum combining TAFTS and ARIES measurements. These will be referred to as fixed temperature retrievals and are shown in figure 4.15.

As with the original ARIES water vapour retrieval, the ARIES fixed temperature water vapour

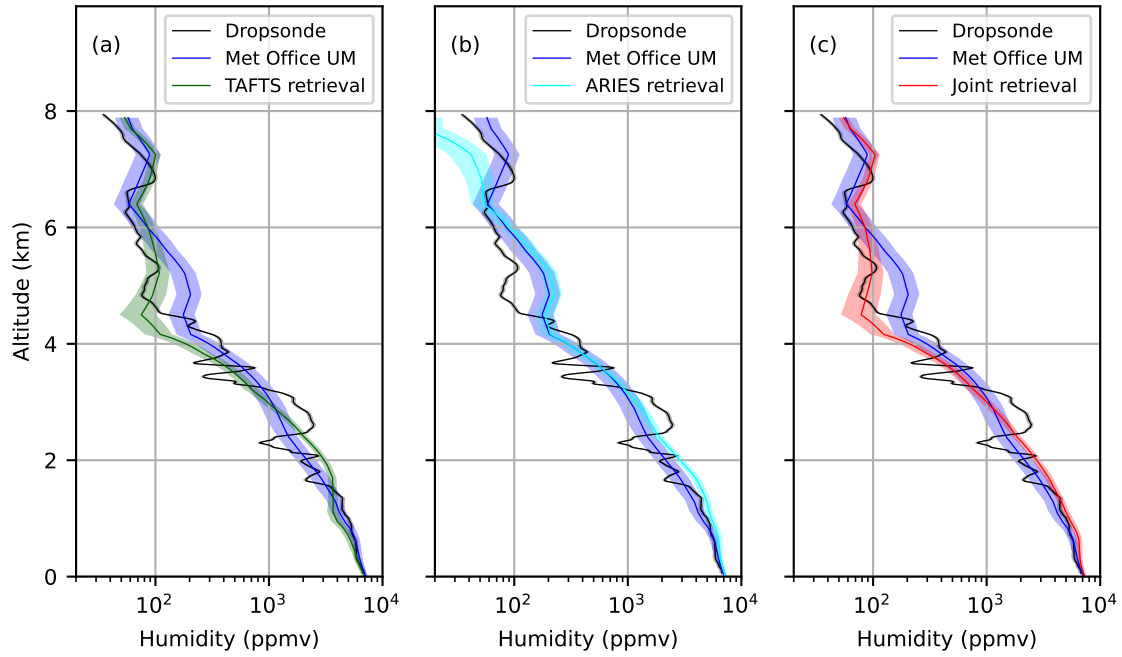


Figure 4.15: Water vapour only retrievals for (a) reduced range TAFTS (b) ARIES and (c) a joint retrieval. The a-priori and sonde 8 profiles are also shown. The shading around each line has the same meaning as in figure 4.11

retrieval shows little improvement over the a-priori water vapour profile and is significantly drier than the sonde above 6.5 km. The TAFTS fixed temperature retrieval matches the sonde profile more closely than the ARIES fixed temperature retrieval, as indicated by the lower RMSE values, see table 4.5, however it is also drier than the TAFTS reduced range retrieval (figure 4.12) in the 4-5 km region and this is reflected in the slightly higher RMSE values in this region and also throughout the profile. This increase in RMSE values indicates that in this case the retrieved water vapour profile has to move slightly further away from the sonde values to compensate for the effects of using the sonde temperature profile. Combining TAFTS and ARIES in the joint retrieval significantly reduces RMSE values relative to the ARIES only case across the majority of the profile. The RMSE values are also slightly reduced from the TAFTS only case.

The joint retrieval has the highest number of DFS (table 4.6) but this value is only a slight improvement over the TAFTS only case. Overall, these results reinforce the beneficial impact

that the measurement of the FIR radiances has for the retrieval of water vapour for this case study, particularly in the mid-upper troposphere.

RMSE	Water Vapour ($\log(\text{kg/kg})$)			
	UM	TAFTS	ARIES	Joint
Overall	0.42	0.39	0.51	0.37
0 – 2 km	0.11	0.23	0.26	0.21
2 – 4 km	0.50	0.51	0.50	0.49
4 – 6 km	0.63	0.42	0.72	0.36
6 – 8 km	0.31	0.35	0.55	0.38

Table 4.5: RMSE values for the UM, TAFTS (reduced range), ARIES and joint fixed temperature retrievals.

DFS	$\text{H}_2\text{O}(z)$	T_{skin}
TAFTS (Reduced Range)	6.460	0.049
ARIES	3.551	1.000
Joint (TAFTS Reduced range)	6.654	1.000

Table 4.6: DFS values for water vapour profile and surface skin temperature for the TAFTS (reduced range), ARIES and joint fixed temperature retrievals.

4.2.6 Atmospheric Variability

In this work I assumed that the in-situ sonde profile represented the true state of the atmosphere as observed by TAFTS and ARIES. However, sonde 8 drifted a total of 17 km south-east over approximately 10 minutes after it was released from the aircraft. The aircraft also travelled 250 m while the spectra were taken.

To assess the variability of the atmosphere I examined the water vapour and temperature profiles for all sondes released during the flight. These are shown in figure 4.16. Sondes 1 and 2 are closest to sonde 8 spatially but were released around an hour prior. Sonde 7 was released 9 minutes before sonde 8 but over 60 km away. All sondes measure a drier atmosphere than is predicted by the UM between 5 and 6 km suggesting that the drier atmosphere retrieved

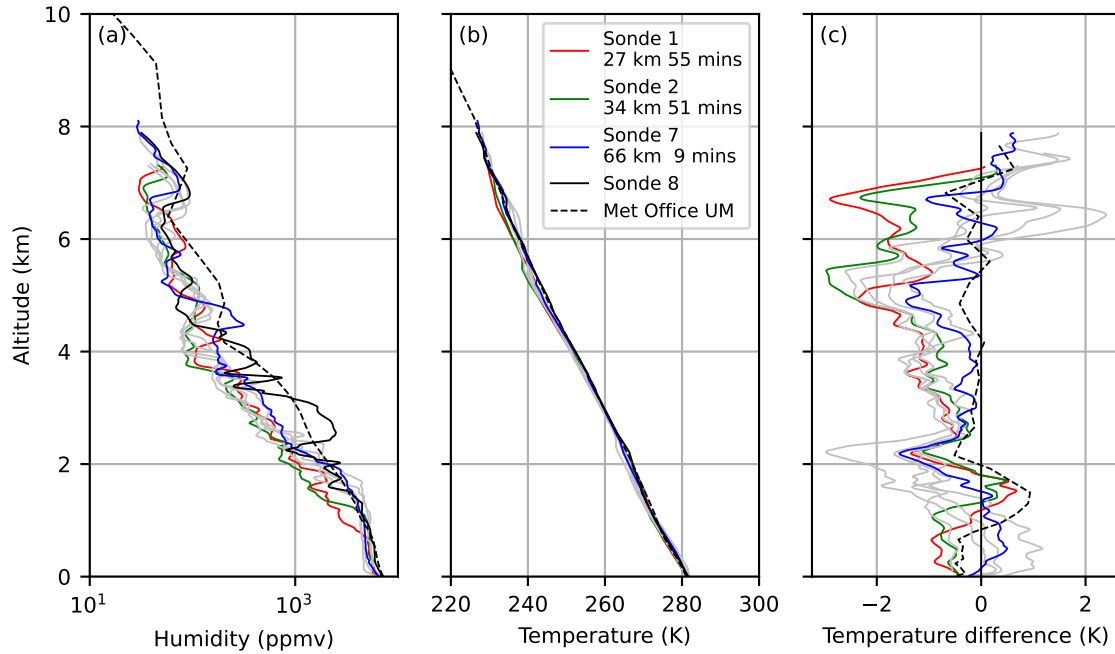


Figure 4.16: All sonde measurements for (a) water vapour and (b) temperature from flight C153. The temperature difference from sonde 8 is shown in (c). The closest sondes in either time or location to sonde 8 are shown in red, green and blue with the other sondes shown in grey. The UM profile used throughout this chapter is also shown as a dashed line.

by TAFTS is representative of the true atmospheric profile. Between 2-3 km, the moist layer measured by sonde 8 is not seen by the other sondes. There is thus the possibility that this wet layer is very localised such that the atmosphere observed by TAFTS and ARIES is drier than suggested by the sonde in this region. Although this layer in the atmosphere is sounded by the TAFTS wavenumbers between 425 and 550 cm^{-1} (see figure 4.5) where the negative bias has been observed in the forward modelling, the lack of similar bias in the ARIES measurements that also sound this layer suggests that this possibly localised wet layer is not the cause of the bias seen between 425 and 550 cm^{-1} .

For the temperature measurements, profiles 1 and 2 are cooler than profiles 7 and 8 above 3 km, suggesting that the atmosphere in this region warmed slightly over the course of an hour. The difference between the profiles measured by sondes 7 and 8 is at most 1.5 K. Taking this as an upper estimate of atmospheric variability, then the temperature deviations seen below

2km in the full range TAFTS retrieval (figure 4.11) and above 7km in the ARIES temperature and water vapour retrieval (figure 4.12) cannot be explained solely by atmospheric variability.

4.3 Conclusions

Overall the forward modelling and retrievals carried out in this section confirm observationally the sensitivity of the FIR to atmospheric water vapour.

Forward modelling highlighted possible issues with the TAFTS spectra at wavenumbers above 425 cm^{-1} that could not be explained by uncertainty in surface properties or spectroscopy. Also a bias in the ARIES measurements of around 3 K in the 667 cm^{-1} CO₂ band was identified.

This information aided the interpretation of the retrievals performed using TAFTS and ARIES radiances. These retrievals found higher DFS and lower RMSE values for both water vapour and temperature for the TAFTS reduced range retrieval compared to the ARIES retrieval. This advantage was maintained for water vapour even when the TAFTS resolution was decreased and the ARIES calibration error was decreased.

This study is the first to use observed aircraft radiances to retrieve water vapour profiles from FIR radiances, and to practically demonstrate the advantage that the FIR has over the MIR, at least for this instrumental configuration. Further retrievals were also performed in collaboration with the Met Office using a different retrieval code. This work confirmed that the results reported here are robust to the choice of retrieval algorithm and hold across the conditions measured by sondes 6 and 7 as well as 8. These retrievals cannot be discussed in depth as a full investigation using the Met Office code was not possible due to consequences of the COVID pandemic.

Chapter 5

Developing FINESSE

This chapter introduces the FINESSE instrument. FINESSE is designed to make in-situ measurements of surface emissivity in the MIR and FIR. Section 5.1.1 describes the specifications of the instrument, sections 5.1.2 and 5.1.3 explain the calibration process and method for calculating the noise equivalent spectral radiance (NESR) and calibration error. Section 5.1.4 shows the instrument line shape. The chosen method for retrieving emissivity is described in section 5.2. Section 5.3 introduces a simulator that has been written for FINESSE and in section 5.4 this simulator is used to model the emissivity retrieval under the conditions seen in London.

The FINESSE project began at Imperial in 2017 with the aim of developing an instrument to make in-situ measurements of surface emissivity. A Bruker EM 27 spectrometer was purchased with initial funding from the National Centre For Earth Observation. The calibration system was designed and built by Jon Murray with support from the Mechanical Instrumentation workshop at Imperial. The control interface for the interferometer and calibration system was written by Alan Last. When I joined the group in 2018 the instrument was just ready to begin

measurement testing, this formed an early part of my research work which helped inform on best practice for data acquisition and processing and allowed me to familiarise myself with the instrument. The instrument continued to evolve over the course of my PhD and I was involved in this process as part of the wider team. My individual contributions to the project were the development of the calibration and data analysis procedure in Python and writing a simulator for the instrument. An instrument paper describing the FINESSE instrument, its spectral characteristics and the spectral calibration is currently in preparation (Jon Murray et al. in prep).

5.1 Description of FINESSE

5.1.1 The FINESSE Instrument

Figure 5.1 shows several photographs of FINESSE. FINESSE consists of a commercial Bruker EM 27 Fourier transform infrared spectrometer (figure 5.1a) coupled with a custom external calibration and scene view system designed and built at Imperial College (figure 5.1b). The spectrometer has a liquid nitrogen cooled extended mercury cadmium telluride (MCT) detector, potassium bromide (KBr) beamsplitter and diamond optical window resulting in a spectral range of 400 to 1600 cm^{-1} (6.3 to 25 μm). The extended MCT detector and KBr window were options added by Bruker and the diamond window was a subsequent modification made once the instrument was at Imperial. The maximum nominal resolution of the spectrometer is 0.5 cm^{-1} corresponding to an optical path difference of 1.21 cm. FINESSE has a field of view of 38 mm at the input window. This diverges with an angle of 1.7°. The external calibration system consists of an ambient temperature and a heated blackbody cavity, hereafter referred

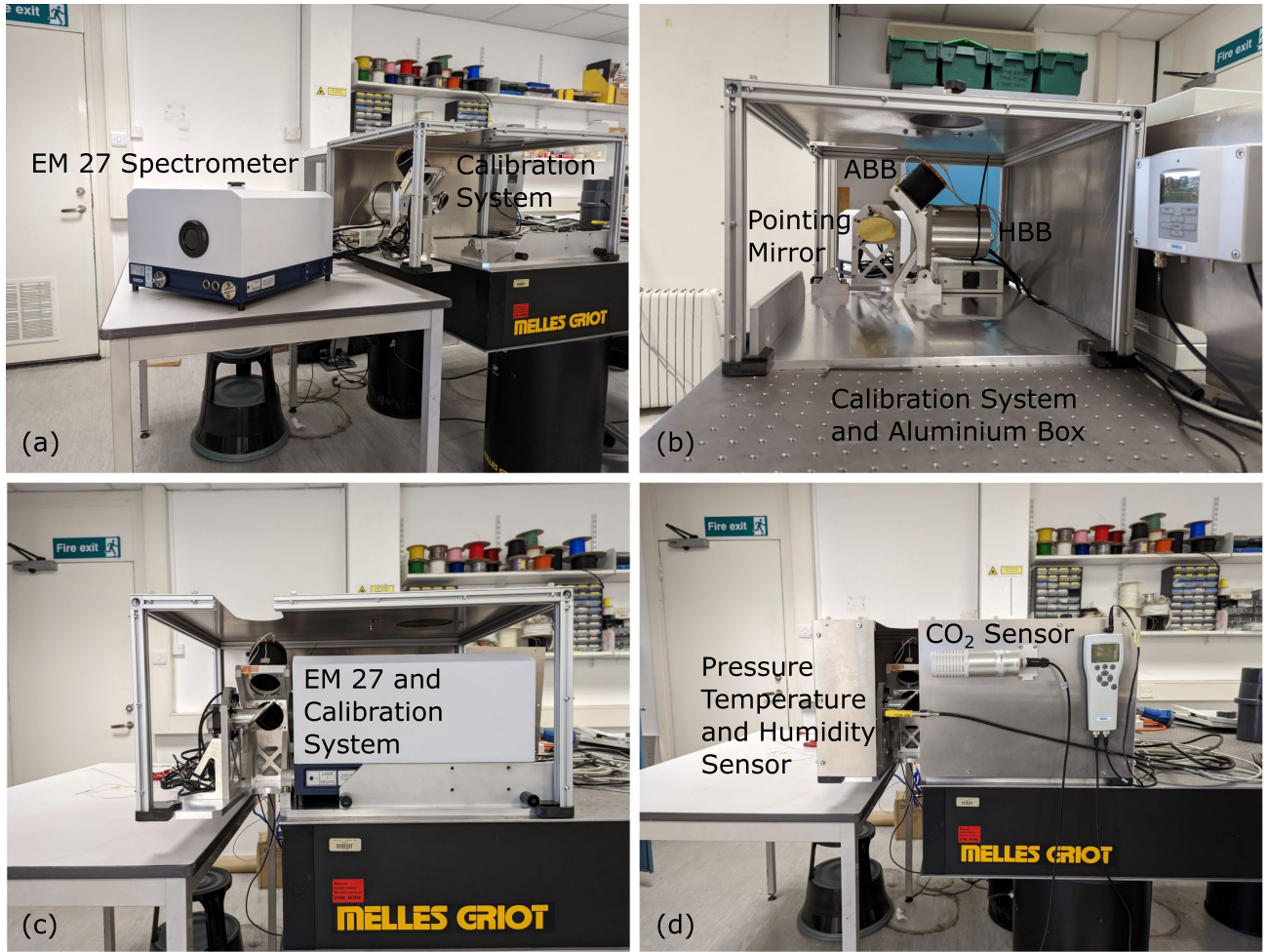


Figure 5.1: Photographs of FINESSE. (a) The Bruker EM 27 and calibration system. (b) Another view of the calibration and pointing system showing the hot black body (HBB) ambient black body (ABB) and pointing mirror. (c) The EM 27 in place within the aluminium box. (d) The aluminium box closed up and the CO₂ and temperature, pressure, humidity sensors in place.

to as ABB and HBB respectively, and a gold pointing mirror. The pointing mirror allows the spectrometer to view the blackbody cavities and external scenes at angles ranging from nadir to zenith. The angle of the pointing mirror and the operation of the EM 27 are controlled by a custom graphical user interface which also monitors the temperatures from the calibration targets. These temperatures are measured using platinum resistance thermometers (PRTs) which are logged using an Omega DP9800. Additional PRTs can be added to monitor extra temperatures as required. The EM 27 and calibration system are enclosed in an aluminium box (figure 5.1d). FINESSE has dimensions 73 x 60 x 36 cm (width x depth x height) and weighs

around 35 kg. The electronics and controllers for the pointing system and heaters are housed in a separate electronics box with dimensions 60 x 56 x 38 cm. The interferograms measured by the EM 27 are accompanied by additional environmental monitoring. This monitoring consists of pressure, temperature and humidity measurements from a Vaisala PTU300 (PTU) and CO₂ measurements from a Vaisala GMP343. These sensors are mounted on the aluminium box which encases the interferometer (figure 5.1d). When making measurements FINESSE will generally cycle through views of the hot and ambient calibration targets and then the scene.

5.1.2 Calibration Process

The most straightforward way to calibrate the spectra produced by FINESSE would be to work in the wavenumber domain, i.e. with the spectra, using the standard Mertz correction described in section 2.1. However, initial measurements of downwelling radiance showed unexpectedly high radiance in the atmospheric window. An example of this is shown in figure 5.2a, which shows a measured downwelling spectrum calibrated using the method described in section 2.1 and a downwelling spectrum simulated with LBLRTM using the atmospheric profile from ERA5 at the closest point in space and time to the measurement. The simulation was also apodised with the FINESSE instrument lineshape.

The measured spectrum is higher than the simulation in the window region and shows two large troughs between 950 and 1000 cm⁻¹ which are due to the absorption of polypropylene (see figure 6.6). Expected features such as emission by ozone just above 1000 cm⁻¹ are not visible in the measured spectrum but are clearly visible in the simulation. Further investigation revealed anomalous behaviour of the phase function similar to that observed by Revercomb et al. (1988) when developing the calibration system for the High-Resolution Interferometer Sounder

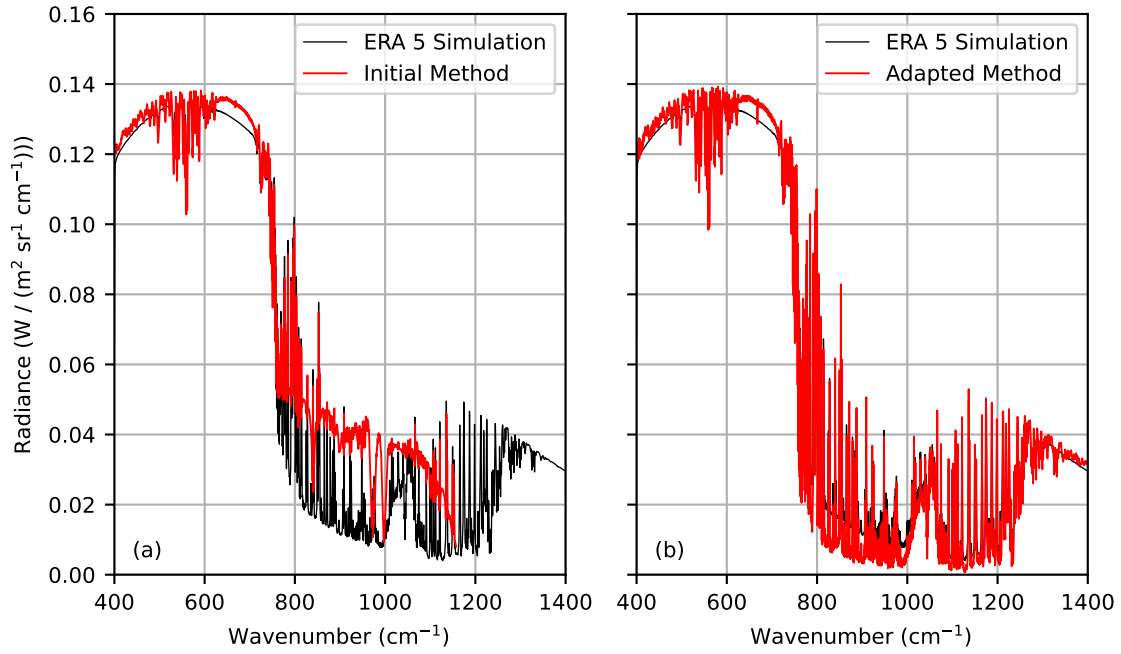


Figure 5.2: (a) Downwelling spectrum calibrated using the method described in section 2.1 in red and the downwelling spectrum simulated using the closest ERA5 profile in space and time in black. (b) The same spectrum calibrated using the method described in section 5.1.2 and the same simulated spectrum.

(Murray personal communication). As Revercomb et al. explain, the standard method, as used in figure 5.2a, only considers the phase component from the sources measured by the interferometer but does not account for radiance from the instrument itself, which can have different phase characteristics. This is particularly important for our purposes as the scene radiance may be low compared to the radiance from the instrument when measuring cold surfaces such as ice or radiation emanating from high in the atmosphere. Therefore, to calculate the response function for FINESSE, we have adapted the method described by Revercomb et al. (1988).

The response function is calculated by first differencing the measured interferograms for the hot and ambient targets

$$Int_{resp} = Int_{HBB} - Int_{ABB}, \quad (5.1)$$

see steps 1 and 2 in figure 5.3.

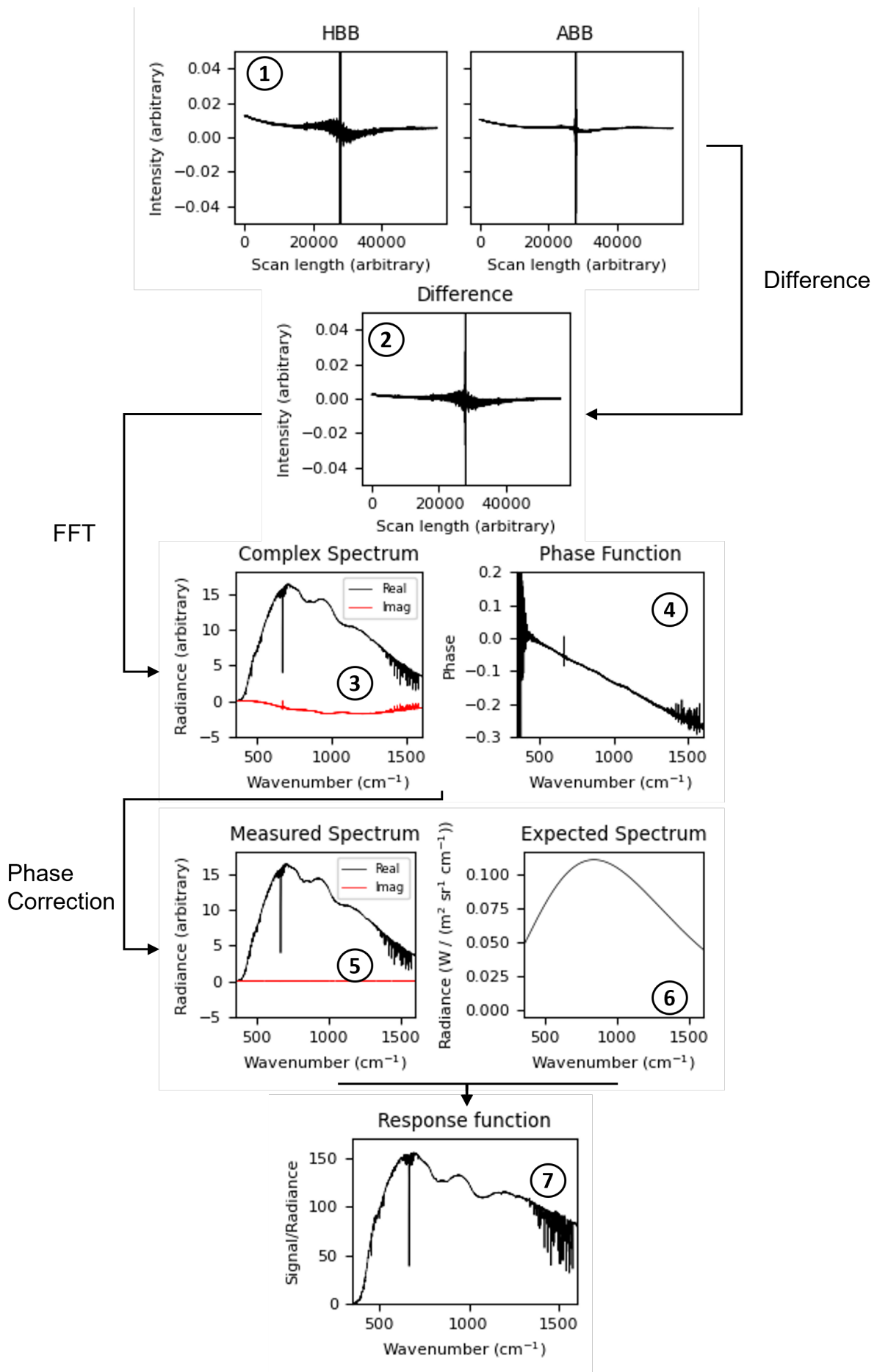


Figure 5.3: A schematic showing how the FINESSE response function is calculated.

A complex Fourier transform is then performed on Int_{resp} to produce a complex difference spectrum $\tilde{S}(\nu)$, (fig 5.3 step 3) where

$$\tilde{S}(\nu) = S_{re}(\nu) + iS_{im}(\nu). \quad (5.2)$$

We then perform a phase correction on the complex difference spectrum. The phase function (fig 5.3 step 4) is calculated using equation 2.8 and is then applied to $\tilde{S}(\nu)$ to produce a phase corrected difference spectrum, $\tilde{S}^{corrected}(\nu)$ (fig 5.3 step 5), where

$$\tilde{S}^{corrected}(\nu) = S_{re}^{corrected}(\nu) + iS_{im}^{corrected}(\nu) \quad (5.3)$$

The real part of this phase corrected difference spectrum, $S_{re}^{corrected}$ is used to calculate the response function, $R(\nu)$ (fig 5.3 steps 6 and 7),

$$R(\nu) = \frac{S_{re}^{corrected}(\nu)}{[L_{HBB}(\nu, T_{HBB}) - L_{ABB}(\nu, T_{ABB})]} \quad (5.4)$$

where $L_{HBB}(\nu, T_{HBB})$ and $L_{ABB}(\nu, T_{ABB})$ are the wavenumber dependent radiances produced by the HBB and ABB at temperatures, T_{HBB} and T_{ABB} , respectively.

The calibrated scene radiance, $L_{scene}(\nu)$, is found by first differencing the scene and HBB interferograms to produce a difference interferogram, Int_{diff} ,

$$Int_{diff} = Int_{scene} - Int_{HBB}. \quad (5.5)$$

A Fourier transform is applied to this difference interferogram and the resulting spectrum is then phase corrected. This produces a phase corrected but uncalibrated complex scene view

spectrum,

$$\tilde{S}^{scene}(\nu) = S_{real}^{scene} + iS_{im}^{scene}. \quad (5.6)$$

Finally, the calibrated scene radiance, $L_{scene}(\nu)$, is calculated using the response function,

$$L_{scene}(\nu) = \frac{S_{real}^{scene}(\nu)}{R(\nu)} + L_{HBB}(\nu, T_{HBB}). \quad (5.7)$$

As mentioned in section 2.1 we must also adjust the wavenumber scale to account for a slight misalignment between the laser and infrared beam and the divergence of the infrared beam. This stretch to the wavenumber scale was determined by the comparison of transmission spectra recorded by the instrument with LBLRTM simulations (Murray, Personal Communication, 2022). The wavenumber multiplier was found to be 1.00016 or 0.016% of the quoted wavenumber scale, which, although small, could be seen at the edges of the sharp water vapour lines. Figure 5.2b shows the same spectrum as figure 5.2a but calibrated using the improved method illustrated in figure 5.3. The spectrum no longer suffers from large polypropylene troughs and shows expected features in the window region such as the ozone band. There is still a slight difference between the simulated and measured spectra between the sharp lines in the atmospheric window. This indicates that the temperature of the simulation higher in the atmosphere did not exactly match the atmosphere viewed by FINESSE. However this comparison still shows the improvement due to the change in calibration method.

5.1.3 Determination of NESR and Calibration Uncertainty

The NESR for the measurements is calculated using the method developed for TAFTS (Green, 2003). This is done using the imaginary part of $\tilde{S}^{scene}(\nu)$. First this is converted into radiance

units using the response function

$$L_{im}(\nu) = \frac{S_{im}^{scene}(\nu)}{R(\nu)}. \quad (5.8)$$

The standard deviation of $L_{im}(\nu)$ is then calculated across a rolling window to produce the NESR as a function of wavenumber. For FINESSE we use a window size of 20 sample points.

The calibration uncertainty is determined by considering the accuracy of the PRTs used to measure the temperatures of the HBB and ABB. This accuracy is 0.2K. To calculate the calibration uncertainty, three versions of the calibrated spectra are produced. The original spectrum is calculated using the measured temperatures of the HBB and ABB. The second spectrum is an upper radiance bound which is calculated using a positive offset of 0.2K to the ABB temperature and a negative offset of 0.2K to the HBB temperature. The third spectrum is as a lower radiance bound calculated using a positive offset to the HBB temperature and a negative offset to the ABB temperature. The difference between the original spectrum and the upper and lower spectra are taken as the upper and lower values of the calibration uncertainty.

Figure 5.4 shows the calculated NESR and calibration error for four example spectra. The four example spectra are views of the upwelling radiance from a water surface taken during the summer and the winter and views of the downwelling radiance from the sky during the summer and winter. The calibration uncertainty is generally larger than the NESR. The calibration uncertainty for the downwelling spectra is significantly higher than that of the upwelling spectra between 800 and 1200 cm^{-1} . This difference is due to the fact that the brightness temperature in this region of the spectrum is significantly lower than the temperature of the ABB calibration target. This is more pronounced for the summer measurement as the ABB target was warmer during the summer measurements at a temperature of 30°C compared to 13°C for the winter

measurements.

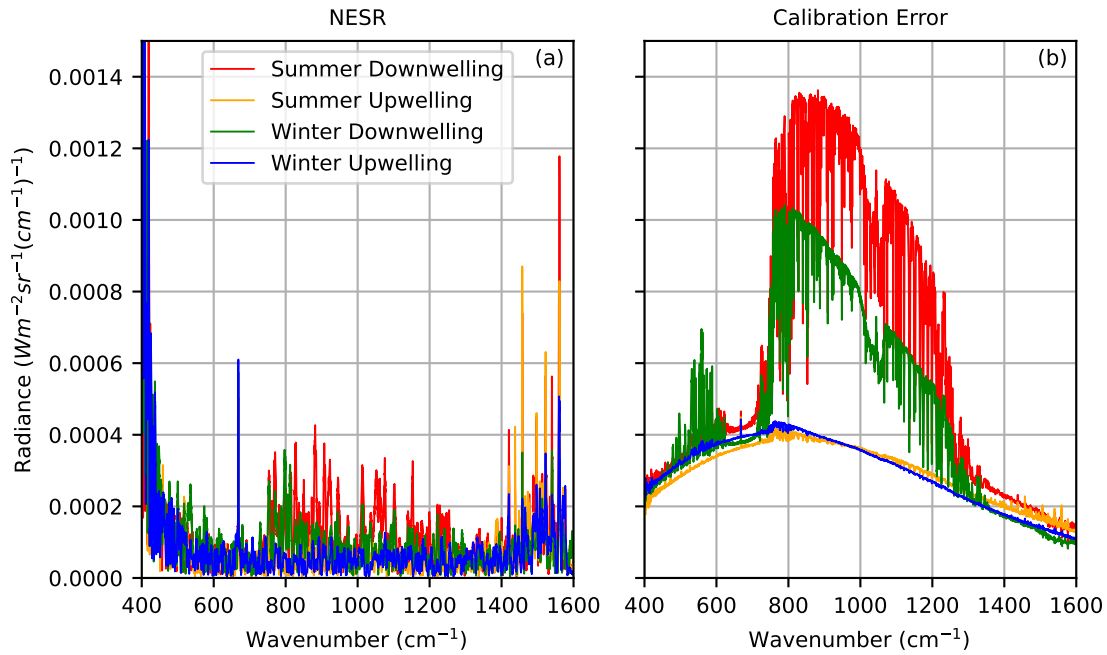


Figure 5.4: The (a) NESR and (b) calibration uncertainty for four example spectra. These spectra consist of views of upwelling radiance from a water surface taken during the summer and the winter and views of the downwelling radiance from the sky during the summer and winter.

5.1.4 Instrument Line Shape

The instrument line shape (ILS) was determined at the same time as the wavenumber stretch factor by comparison between the sharp emission features present in transmission spectra recorded at a nominal resolution of 0.5 cm^{-1} and high-resolution simulations carried out using LBLRTM (Murray, personal communication, 2022). Figure 5.5 shows the ILS as a function of wavenumber.

The ILS widens as the wavenumber of the radiation increases; it also shows some significant asymmetry at higher wavenumbers. The ILS has been modelled by convolving three components. Firstly, a sinc function which is a consequence of the finite 1.21 cm optical path difference of the EM 27 (Griffiths & de Haseth, 1986). Secondly a sinc^2 component, which describes the

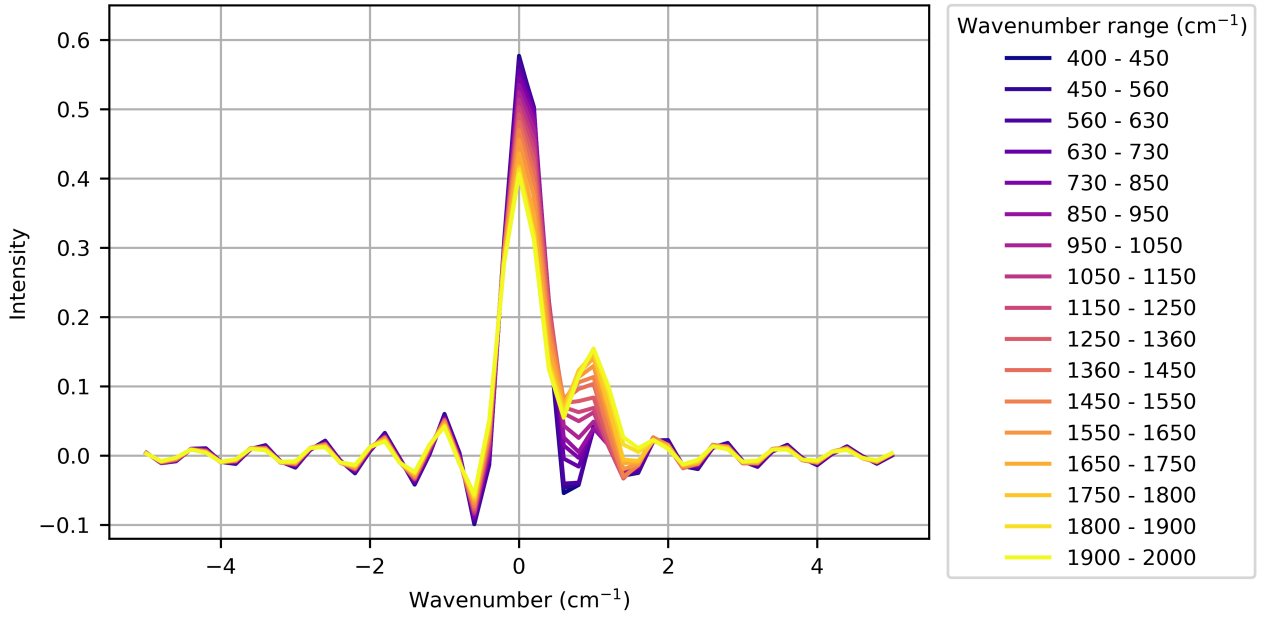


Figure 5.5: The FINESSE instrument line shape as a function of wavenumber.

self apodisation mentioned in section 2.1. The magnitude of this component is dependent on the solid angle of the radiation propagating through FINESSE (Bianchini et al., 2019) which through comparison with simulation was determined to be 0.001 sr. Finally, the asymmetry is modelled following the method of Genest and Tremblay (1999) who describe a mathematical expression for the instrument line shape of a Fourier Transform spectrometer with the detector situated off the optical axis. The parameters for the instrument line shape were derived empirically by applying the ILS to the LBLRTM simulation and comparing this to the measured spectrum. To simplify the application of the instrument line shape, it has been specified for 15 wavenumber intervals, although in reality it changes continuously with wavenumber.

5.2 Emissivity Retrieval Method Chosen

We have chosen to adapt an emissivity retrieval method that has been successfully applied for the retrieval of surface emissivity in the MIR using high resolution spectra (Newman et al.,

2005). This method was used to retrieve surface emissivity of water at different temperatures and salinities using observations from ARIES taken from aircraft and in the lab and the results agreed well with theoretical emissivity calculations. It has also been adapted for the retrieval of FIR snow and ice emissivity from aircraft measurements (Bellisario et al., 2017). The method involves the measurement of upwelling radiation from the surface under study, as well as direct measurement of the downwelling radiation and determination of the transmission between the surface under study and the detector. Figure 5.6 shows the geometry of this measurement technique.

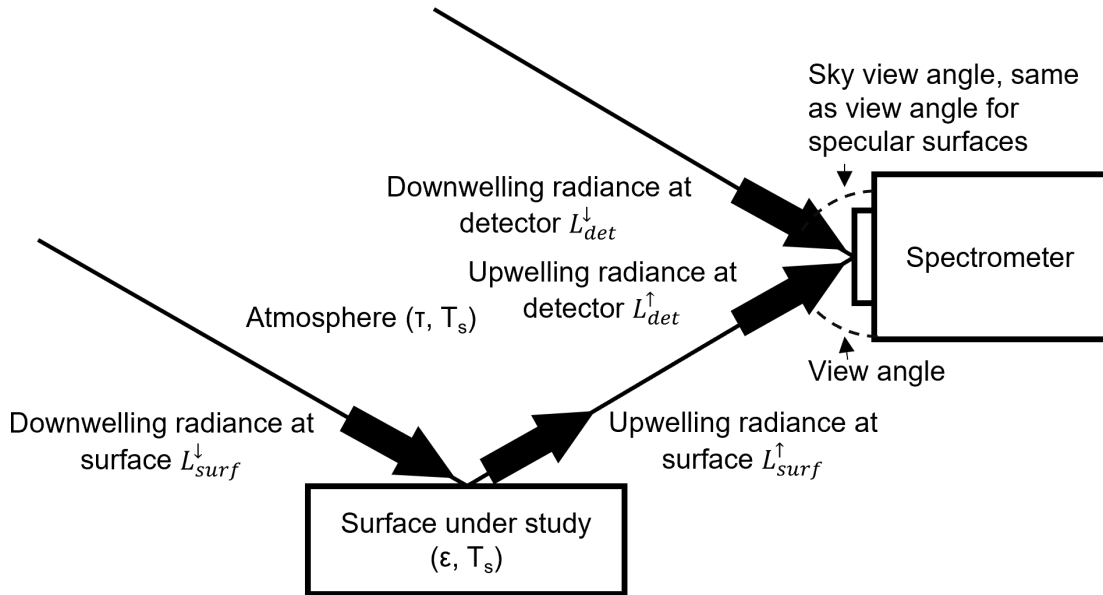


Figure 5.6: A schematic showing the geometry of the chosen emissivity retrieval method.

Selecting this method allows us to make in-situ measurements without disturbing or heating the surface being measured. It is immediately clear why heating the surface is not appropriate for snow and ice measurements. Making in-situ measurements of snow and ice surfaces is important as the properties of the snow such as grain size and shape can vary quickly throughout the day and can be altered by transporting the snow. Furthermore, the emissivity of snow and ice is dependent on these properties (Hori et al., 2006). Being able to make in-situ measurements is also important for measuring the emissivity of other natural surfaces. For example the

emissivity of soil samples has been shown to differ between field and lab measurements due to changes in soil moisture when the sample is transported (Mira et al., 2009). Furthermore, the emissivity of in-homogeneous surfaces, such as a mixture of sand and gravel, cannot simply be reproduced by combining laboratory measurements of pure surface types (Göttsche et al., 2018). We also measure the downwelling radiance directly as opposed to making a reflectance measurement from a gold plate as in for example Hori et al. (2006) or Horton et al. (1998). This is because it was easy for us to make direct sky measurements due to the FINESSE pointing system however work should be done to examine the performance of different emissivity retrieval techniques including the use of a gold plate.

Following the derivation by Newman et al. (2005), the upwelling radiance from a surface, $L_{surf}^{\uparrow}$ which is a function of viewing angle θ , is composed of a thermal emission term and a component of reflected radiation both of which are dependent on the emissivity,

$$L_{surf}^{\uparrow}(\theta) = \epsilon(\theta)B(T_s) + (1 - \epsilon(\theta))L_{surf}^{\downarrow}(\theta), \quad (5.9)$$

where ϵ is the emissivity, $L_{surf}^{\downarrow}$ is the downwelling radiation at the surface and $B(T_s)$ is the Planck function at surface temperature T_s . $L_{surf}^{\downarrow}$ and ϵ are both functions of viewing angle and all values are spectrally dependant. However, when measuring surface emissivity, we must also consider the absorption and emission of the atmosphere between the surface and the detector. Therefore, the upwelling radiance measured by the detector, $L_{det}^{\uparrow}(\theta)$, becomes,

$$L_{det}^{\uparrow}(\theta) = \tau(\theta)L_{surf}^{\uparrow}(\theta) + E^{\uparrow}(\theta), \quad (5.10)$$

where τ is the transmission and $E^{\uparrow}$ the upwelling emission of the atmospheric layer between the surface and the spectrometer. Similarly, the downwelling radiation at the surface differs

from that measured by the spectrometer,

$$L_{surf}^{\downarrow}(\theta) = \tau(\theta)L_{det}^{\downarrow}(\theta) + E^{\downarrow}(\theta), \quad (5.11)$$

where $E^{\downarrow}$ is the downwelling emission of the layer between the detector and the surface. During our in-situ measurements the distance between the spectrometer and the surface is at most a few metres, and usually less than 0.5 m, so for these distances we assume that the atmosphere is homogeneous and isothermal, in which case the up- and downwelling emission is the same and can be written as

$$(1 - \tau(\theta))B(T_a) \quad (5.12)$$

where $B(T_a)$ is the Planck function at the average temperature of the atmospheric layer, T_a .

By rearranging the above equations, we find the following expression for surface emissivity,

$$\epsilon = \frac{L_{det}^{\uparrow} - \tau^2 L_{det}^{\downarrow} - (1 - \tau^2)B(T_a)}{\tau\{B(T_s) - \tau L_{det}^{\downarrow} - (1 - \tau)B(T_a)\}}. \quad (5.13)$$

Accurately and precisely determining the surface skin temperature is a vital part of the retrieval process. Many algorithms have been developed to do this when determining emissivity in the MIR (Salvaggio & Miller, 2001). However many of these techniques make the assumption that the transmission between the surface and the detector is 1, which is not appropriate for the FIR where there is strong water vapour absorption. Therefore I decided to use the surface temperature retrieval method described by Newman et al. (2005) which relies on the spectral smoothness of the surface emission but takes into account the atmospheric path between the surface and the instrument. This is done by noting that $L_{surf}^{\uparrow}$ is composed of emitted surface radiation, which is a smoothly varying function, and reflected sky radiation which has spectral

features. Therefore it should be possible to find a value for the reflectance, ρ , such that

$$L_{surf}^{\uparrow} - \rho L_{surf}^{\downarrow} = \epsilon B(T_s) \quad (5.14)$$

is a smooth function. By splitting equation 5.14 into small wavenumber intervals (Newman et al. use 40 cm^{-1}) ρ can be taken as constant within each wavenumber interval. Substituting in definitions from equations 5.10, 5.11 and 5.12, equation 5.14 can be rewritten as

$$\frac{1}{\tau} \{L_{det}^{\uparrow} - (1 - \tau)B(T_a)\} - \rho \{\tau L_{det}^{\downarrow} + (1 - \tau)B(T_a)\} = \epsilon B(T_s). \quad (5.15)$$

As this equation should still represent a smooth function, the value of ρ can be altered in each wavenumber interval to minimise the root mean square difference between the left hand side of the equation and a quadratic fit of the left hand side of the equation. Figure 5.12 illustrates this process in action. The average emissivity for the wavenumber band is $1 - \rho$. Therefore the right hand side of the equation can be used to determine the surface temperature by inverting the Planck function.

5.3 The FINESSE Simulator

The aim of the FINESSE simulator was to create a computational tool to investigate how the emissivity measurement will perform under different environmental conditions and with different measurement uncertainties. As detailed in section 5.2 the emissivity calculation is based on the method used by Newman et al. (2005) with emissivity calculated using equation 5.13. The simulator is written predominantly in Python, however the calculation of the downwelling

spectrum and the transmission require the use of LBLRTM and must therefore be performed on a Linux system. Figure 5.7 is a schematic showing how the simulator works. The inputs to the simulator are in blue. These consist of:

- Atmospheric profile above FINESSE or measured downwelling spectrum
- Near surface air temperature, humidity, CO₂ and pressure
- Path length between surface and FINESSE
- Theoretical emissivity values from the Huang et al. (2016) dataset
- Surface temperature
- ILS
- Measurement uncertainties as desired including
 - NESR
 - Calibration uncertainty
 - Path length
 - Near surface air temperature, humidity, CO₂ and pressure

If a measured downwelling spectrum is not provided, the atmospheric profile is used to calculate the unperturbed downwelling spectrum at the detector at a resolution of 0.001 cm^{-1} using LBLRTM. The unperturbed transmission is also calculated at a resolution of 0.001 cm^{-1} using LBLRTM in horizontal mode. The simulations are then convolved with the ILS. Finally, the unperturbed upwelling spectrum at the detector, $L_{det}^{\uparrow}$, is calculated by rearranging equation 5.13

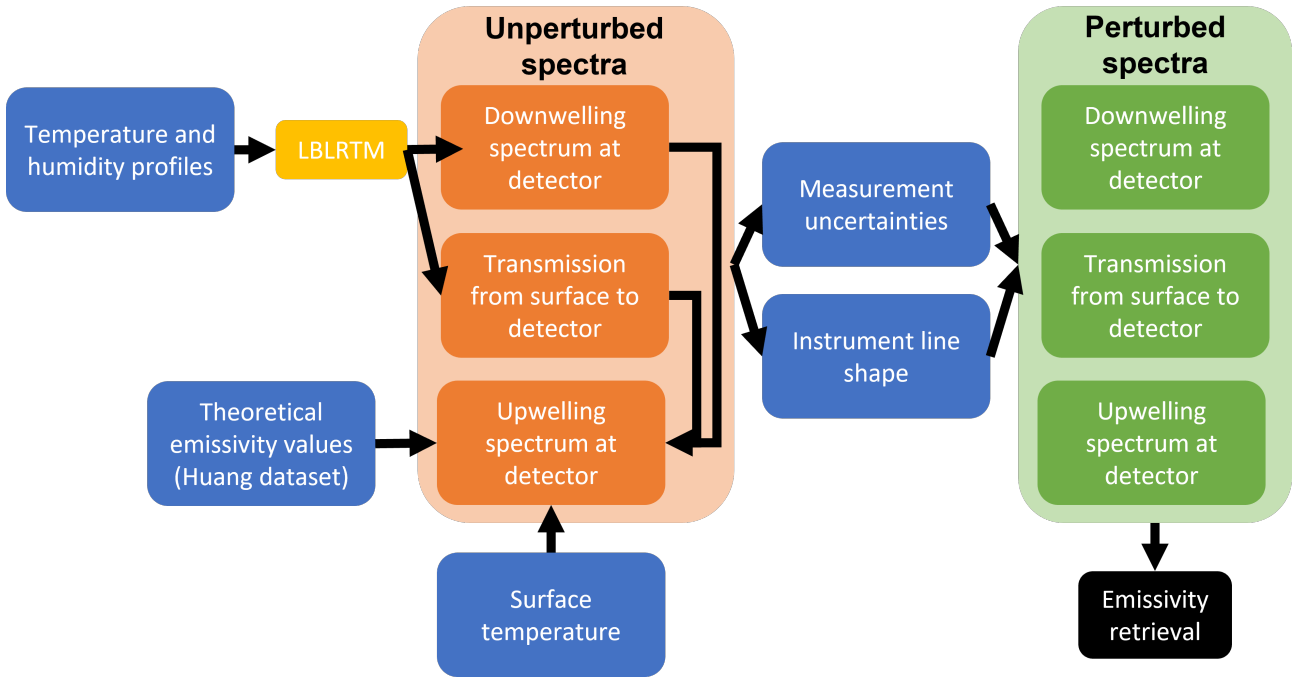


Figure 5.7: Diagram showing the FINESSE simulator

to give

$$\begin{aligned}
 L_{det}^{\uparrow} &= \epsilon \tau B(T_s) && \text{surface emission} \\
 &+ (1 - \epsilon) \tau^2 L_{det}^{\downarrow} && \text{reflected downwelling radiation} \\
 &+ (1 - \tau) B(T_a) && \text{direct atmospheric emission} \\
 &+ (1 - \epsilon) \tau (1 - \tau) B(T_a) && \text{reflected atmospheric emission}
 \end{aligned} \tag{5.16}$$

where ϵ is the emissivity from the Huang dataset, τ the previously calculated unperturbed transmission, $B(T_s)$ the Planck function at temperature T_s of the surface, $L_{det}^{\downarrow}$ the unperturbed downwelling radiance at the detector and $B(T_a)$ the Planck function at temperature T_a of the atmosphere. All variables other than temperature are functions of wavenumber and angle. Perturbations can then be applied to the unperturbed spectra and transmission to simulate the measured spectra. The emissivity can then be calculated and compared to the Huang emissivity.

5.4 Modelled Emissivity Retrievals

The aim of this section was to use the FINESSE simulator to investigate the performance of the emissivity retrieval under the conditions that were seen in the subsequent emissivity measurements described in detail in section 6.1.

5.4.1 Simulating Unperturbed Spectra

For these tests two sets of atmospheric conditions were used. One set was taken from measurements made on 8-Sep-21, hereafter referred to as the summer case, and the second set from measurements made on 17-Mar-22, the winter case. For more details about these measurements see section 6.1 and table 6.1. Two different surface temperatures were used with each set of atmospheric conditions. One surface temperature was 2K warmer than the atmospheric temperature and one surface temperature was 1K colder. These temperature contrasts were representative of the conditions observed during the measurements analysed later in this thesis. This resulted in four test cases: summer warm surface, summer cold surface, winter warm surface and winter cold surface.

Table 5.1 shows the parameters used for these four models. Although the relative humidity is higher in the winter case compared to the summer case, the winter water vapour concentration of 0.7×10^4 ppmv is less than half the summer amount of 1.5×10^4 ppmv. For all cases the ILS described in section 5.1.4 was applied to the unperturbed transmission spectra. The emissivity spectrum for pure water from Huang et al. (2016) was used as the “true” emissivity of the surface. The aim of the experiment is to test how well this “true” emissivity can be retrieved in different conditions and with different perturbations applied.

Model	Summer	Winter
Downwelling spectrum	Taken at 1151 on 8-Sep-21	Taken at 0906 on 17-Mar-22
Near surface air temperature ($^{\circ}\text{C}$)	26.13	7.03
Near surface humidity (%RH)	44.93	66.59
Near surface CO_2 (ppm)	442	470
Near surface pressure (hPa)	1010	1028
Path length (cm)	34	34
Surface temperature ($^{\circ}\text{C}$)	28 / 25	9 / 6

Table 5.1: Inputs used for the FINESSE simulator.

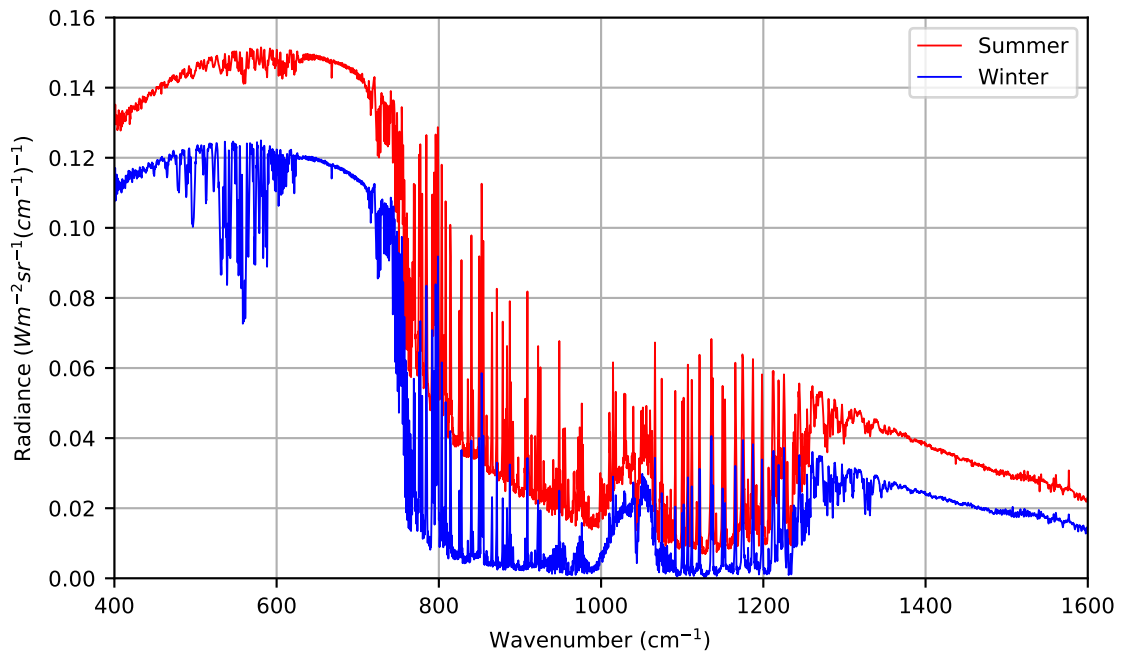


Figure 5.8: The downwelling summer and winter spectra used as inputs to the FINESSE simulator. These spectra are taken from measurements made by FINESSE from the roof of Imperial College.

Figure 5.8 shows the summer and winter downwelling spectra used as input to the FINESSE simulator for these tests. The warmer atmospheric conditions cause higher radiance across the summer spectrum, while in the drier winter conditions the atmosphere is more transparent in the FIR microwindows between 450 and 600 cm^{-1} allowing radiance from higher up in the atmosphere to reach the instrument.

Figure 5.9a shows the unperturbed simulated transmission for the summer case and 5.9b shows the difference between the summer and winter cases. In both cases, the transmission is 1.0 between 700 and 1300 cm^{-1} . In the winter case, the cooler, drier atmospheric layer between the surface and FINESSE leads to a higher transmission both below 600 cm^{-1} and above 1300 cm^{-1} . However in the 667 cm^{-1} CO_2 band, the larger winter CO_2 concentration leads to a lower transmission in the winter case.

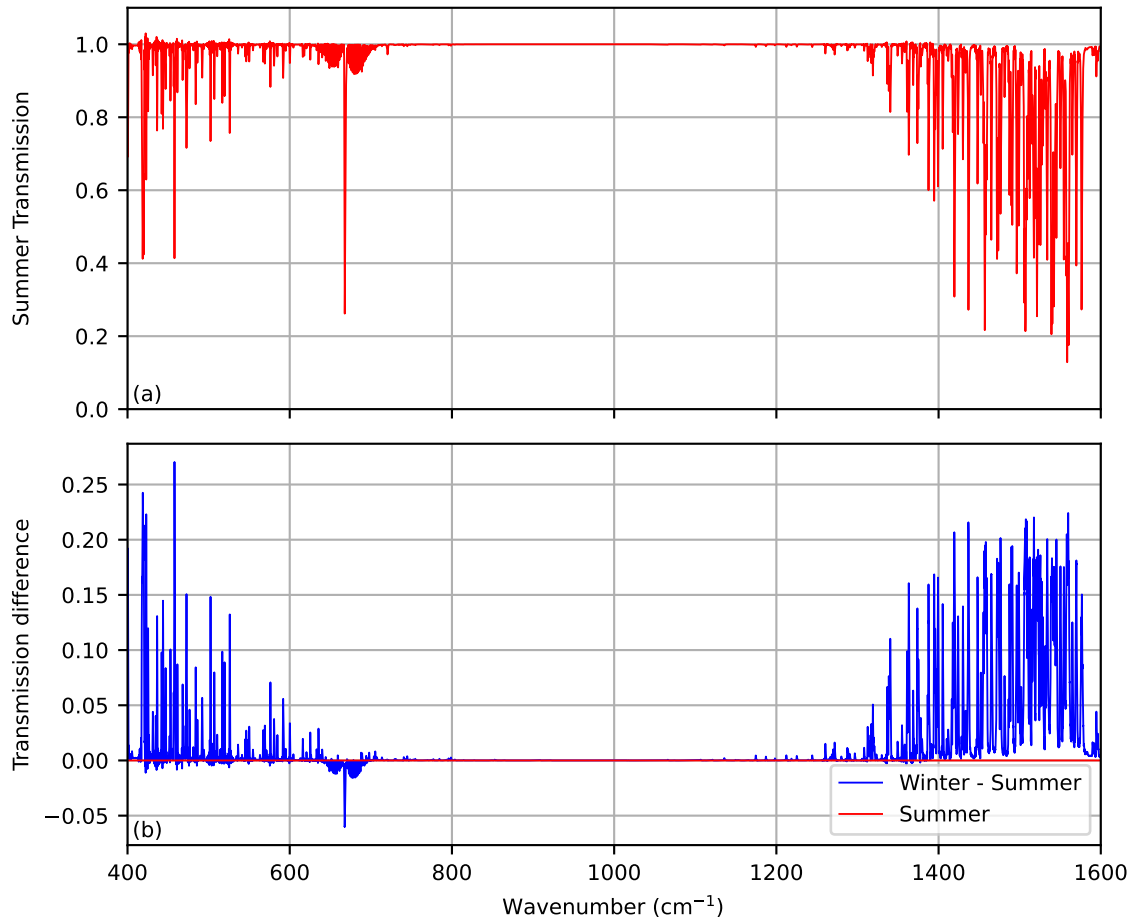


Figure 5.9: (a) The unperturbed simulated transmission between the surface and the detector calculated using the FINESSE simulator for the summer case. (b) The difference in transmission between the winter and summer cases (winter transmission - summer transmission).

Figure 5.10 shows the total unperturbed simulated upwelling spectra at the detector for each of the four cases calculated using equation 5.16. The total spectrum is plotted in black and the individual components of this equation are also plotted in different colours. The radiation

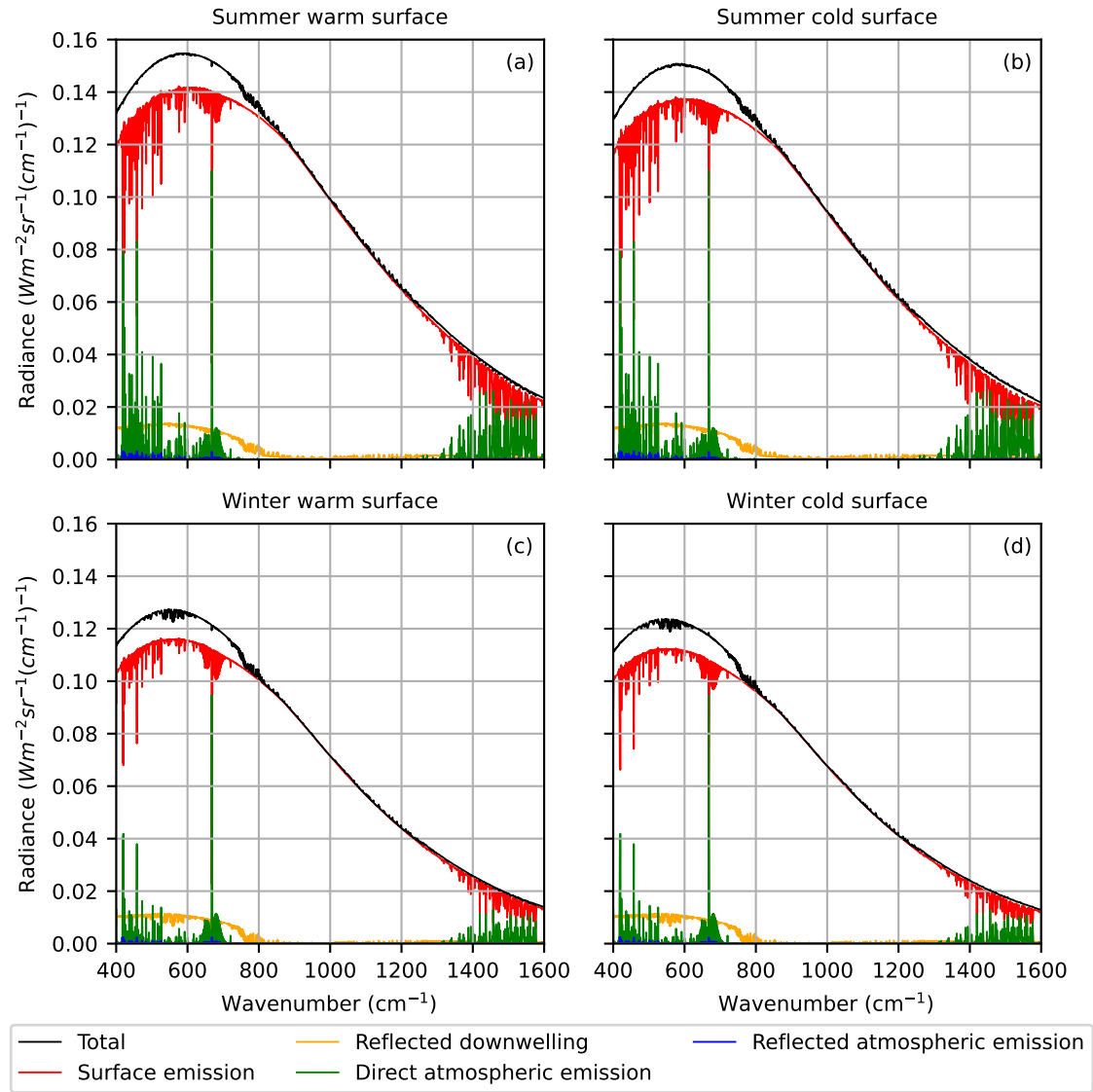


Figure 5.10: The unperturbed simulated upwelling radiation at the detector calculated for the four cases. The total radiation is shown in black and the different colours show the separate terms from equation 5.16.

emitted by the surface dominates the upwelling radiance received at the detector. Between 800 and 1300 cm^{-1} the total upwelling radiance is almost entirely composed of radiation emitted by the surface with the remaining fraction comprised of reflected downwelling radiance. Below 800 cm^{-1} the reflected downwelling radiance makes up a larger portion of the total upwelling radiation at the detector, this correlates with spectral shape of the downwelling spectrum (fig 5.8) and lower emissivity of the water surface in this wavenumber range (see emissivity plotted in figure 1.5 or figure 5.16). In the areas of low atmospheric transmission below 600 cm^{-1}

and above 1400 cm^{-1} the direct emission from the atmosphere increases. The summer cases exhibit a larger contribution from the direct atmospheric emission, due to the lower transmission and higher temperature of the atmospheric layer in the summer case. The reflected atmospheric emission is the smallest component of the total upwelling radiance.

5.4.2 Constructing Perturbed Spectra

The types and magnitudes of the measurement uncertainty considered in this analysis are listed in table 5.2.

Uncertainty	Uncertainty Value	Origin
NESR for upwelling spectrum	See figure 5.4	Calculated using method from section 5.1.3
Calibration uncertainty for upwelling spectrum	See figure 5.4	Calculated using method from section 5.1.3
NESR for downwelling spectrum	See figure 5.4	Calculated using method from section 5.1.3
Calibration uncertainty for downwelling spectrum	See figure 5.4	Calculated using method from section 5.1.3
Distance between surface and detector	1 cm	
Atmospheric temperature	0.3°C	Accuracy of Vaisala PTU300
Atmospheric humidity	$1\% + 0.008 \times \text{reading}$	Accuracy of Vaisala PTU300
Atmospheric pressure	0.15 hPa	Accuracy of Vaisala PTU300
Atmospheric CO_2	$3\text{ ppm} + 0.01 \times \text{reading}$	Accuracy of Vaisala GMP343

Table 5.2: The types and magnitudes of measurement uncertainty considered in this section.

The NESR and calibration error were taken from the examples shown in figure 5.4. To determine the impact of the the NESR and calibration uncertainty on the retrieved emissivity, the unperturbed spectra were offset by the value of the uncertainty to produce perturbed spectra and the emissivity retrieval was run with all other inputs held constant. For example in the case of the upwelling NESR, the upwelling spectrum only was increased by the value of the

NESR and the emissivity retrieval was then carried out.

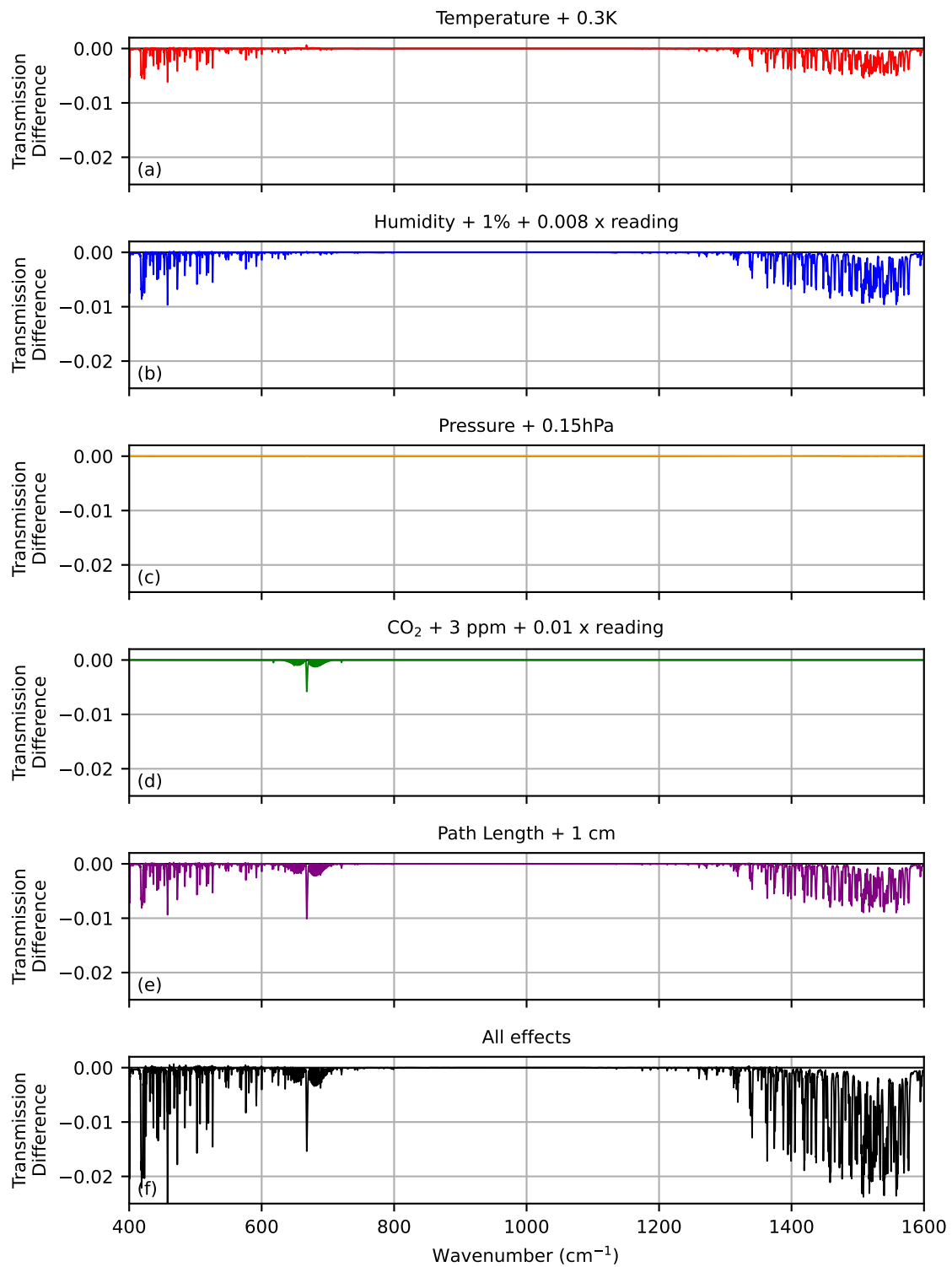


Figure 5.11: The change in transmission caused by the perturbations listed in table 5.2.

The uncertainties in the atmospheric parameters such as humidity and CO_2 concentration

affect the final emissivity because they lead to an error in the simulated transmission between the surface and the instrument. Figure 5.11 shows the change in transmission caused by the specified change in each parameter.

The change in transmission caused by an increase in pressure or CO₂ concentration is very small. The largest changes are caused by a change in the atmospheric humidity or path length. When all perturbations in atmospheric parameters are considered (see figure 5.11f), the largest differences in the transmission are seen below 500 cm⁻¹ and above 1300 cm⁻¹ where the difference caused by the perturbations reaches -0.02. For simplicity, in the rest of the analysis all the atmospheric perturbations were considered together and a maximum and minimum perturbed transmission spectrum was constructed for the summer case and the winter case. The emissivity retrieval was then rerun using the unperturbed upwelling and downwelling spectra but the perturbed transmission spectrum. The atmospheric temperature used in the retrieval equation was also perturbed to be consistent with the perturbed transmission spectrum. For example, for the minimum transmission spectrum the atmospheric temperature used in equation 5.13 was increased by 0.3 K. This is to mimic the case of a measurement error of 0.3 K in the PRT recording the atmospheric temperature causing the measured atmospheric temperature to be higher than the actual atmospheric temperature.

5.4.3 Surface Temperature Retrieval

Figure 5.12 illustrates the process of the Newman surface temperature retrieval for the wavenumber range 1100 to 1140 cm⁻¹ as described in equation 5.15. In this case no perturbations have been applied to the simulation. The solid line for each color is the left side of equation 5.15 at the ρ value indicated in the legend and the dashed line is a quadratic fit of the left side of

equation 5.15 across the interval. The black line is clearly the smoothest and this is confirmed by the lowest RMS value. This means the best estimate for the value of the reflectivity is 0.030 for this wavenumber interval.

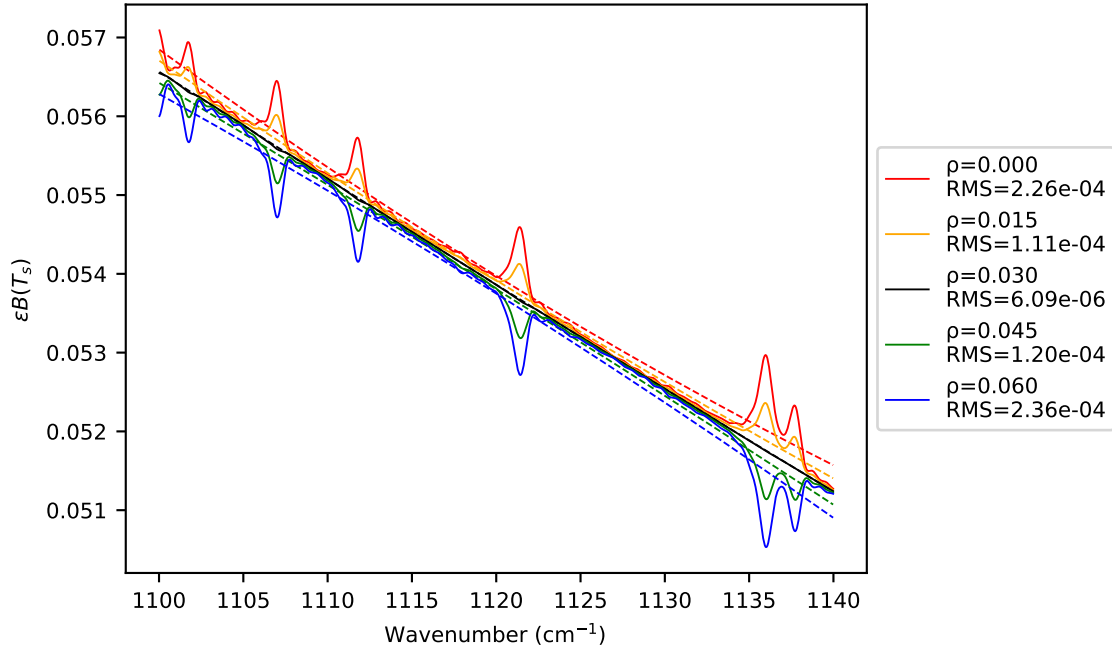


Figure 5.12: Five iterations of the Newman temperature retrieval for the wavenumber range 1100 to 1140 cm^{-1} . A ρ value of 0.030, shown in black minimises the RMS.

The minimisation of the RMS value is carried out using the Python `minimize_scalar` function. The value of the left hand side of equation 5.15 with the best estimate of reflectivity inserted is equal to $\epsilon B(T_s)$. As $\epsilon = 1 - \rho$, the surface temperature can then be found by inverting the Planck function. This process is carried out in 40 cm^{-1} bins.

Figure 5.13 shows the temperature retrieved in each 40 cm^{-1} bin over the FINESSE wavenumber range. The error bars represent the uncertainty from inverting the Planck function. The surface temperature is retrieved as slightly different values in the different wavenumber bins. The retrieved temperature is generally closer to the input temperature in the winter cases. (Note that the y-scale for the summer retrievals covers a range of around 3.5 $^{\circ}\text{C}$ and the y-scale for the winter retrievals covers a range of around 0.25 $^{\circ}\text{C}$). The summer temperature retrieval is

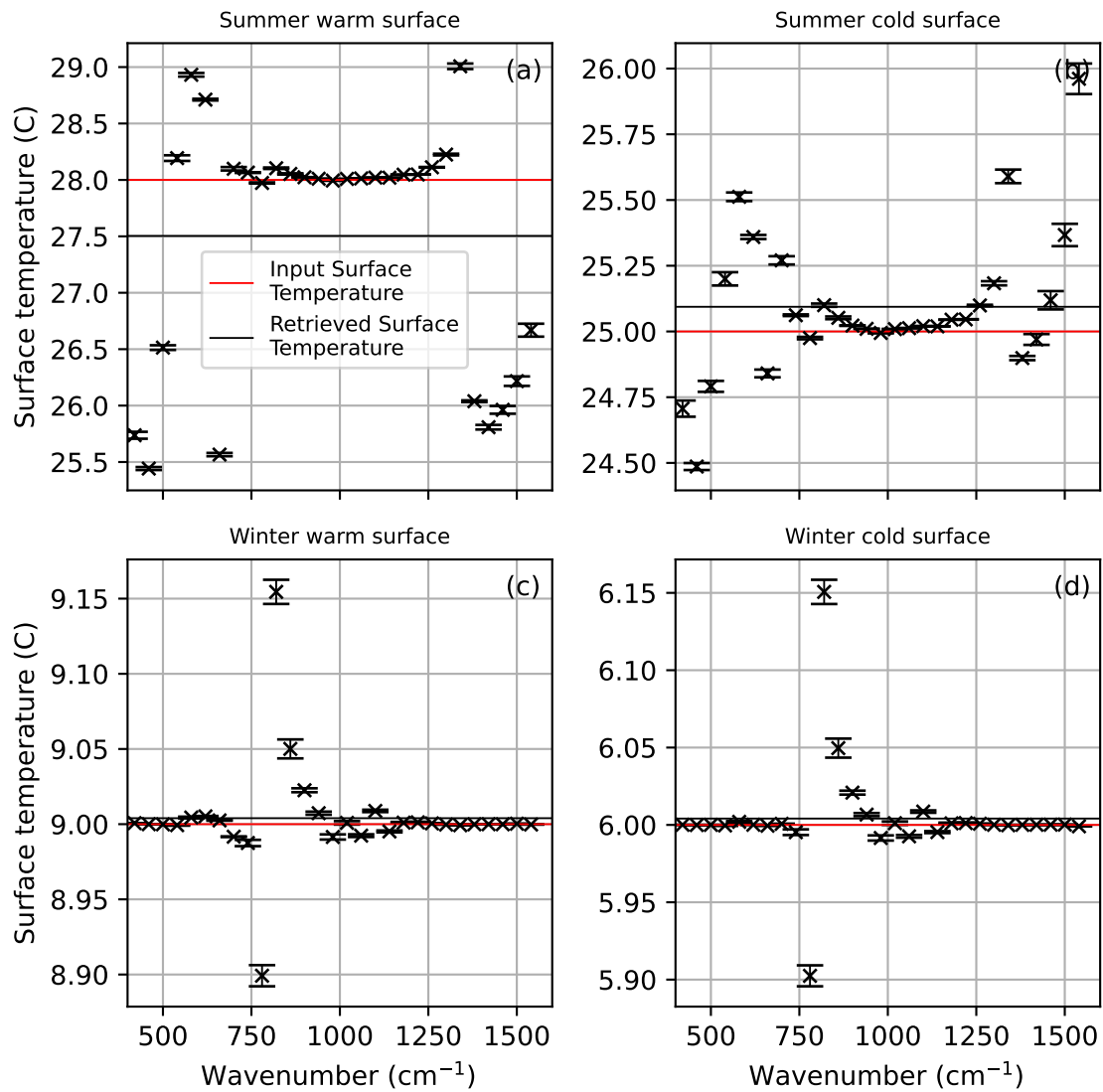


Figure 5.13: The retrieved surface temperature for each 40 cm^{-1} wavenumber bin between 600 and 1600 cm^{-1} . The red horizontal line is the input temperature and the black horizontal line is the average of the retrieved temperature in each bin. The y-scale for the summer retrievals covers a range of around 3.5 °C and the y-scale for the winter retrievals covers a range of around 0.25 °C

worst in regions with strong water vapour absorption however this effect of water vapour is not seen in the winter cases. This could be because the atmosphere is drier in the winter case however more work is needed to thoroughly investigate the temperature retrieval.

Following Newman et al. (2005) I decided to use the wavenumber range 800 to 1200 cm^{-1} for the temperature retrieval. This is justified by the fact that the signal from the surface makes up the largest proportion of the radiance measured by the spectrometer (see figure 5.10). The surface temperature retrieval for this wavenumber range is shown in figure 5.14.

The retrieved surface temperature in all cases is within 0.025°C of the input temperature. The inclusion of the 800 and 840 cm^{-1} wavenumber bins is causing the surface temperature to be slightly overestimated however further work needs to be done to identify the best wavenumber range for the temperature retrieval, particularly as differences can be seen between the behaviour of the retrieval in the summer and winter cases. These temperature retrievals also do not take into account random noise which will reduce the smoothness of the retrieval even at the correct surface temperature. Therefore, pending further investigation with real data, the wavenumber range 800 to 1200 cm^{-1} was chosen to retrieve the temperature.

Figure 5.15 also shows the surface temperature retrievals for the four cases. The black horizontal line is the surface temperature specified in the simulation for each case and the crosses mark the temperature retrievals carried out without perturbations. These unperturbed retrievals are plotted five times for each case with the error bars representing the change in retrieved surface temperature caused by each perturbation to the input spectra and transmission as described in the legend.

In all cases the retrieved surface temperature is within 0.025°C of the surface temperature specified in the simulation with a positive offset observed across all four cases. The offset is

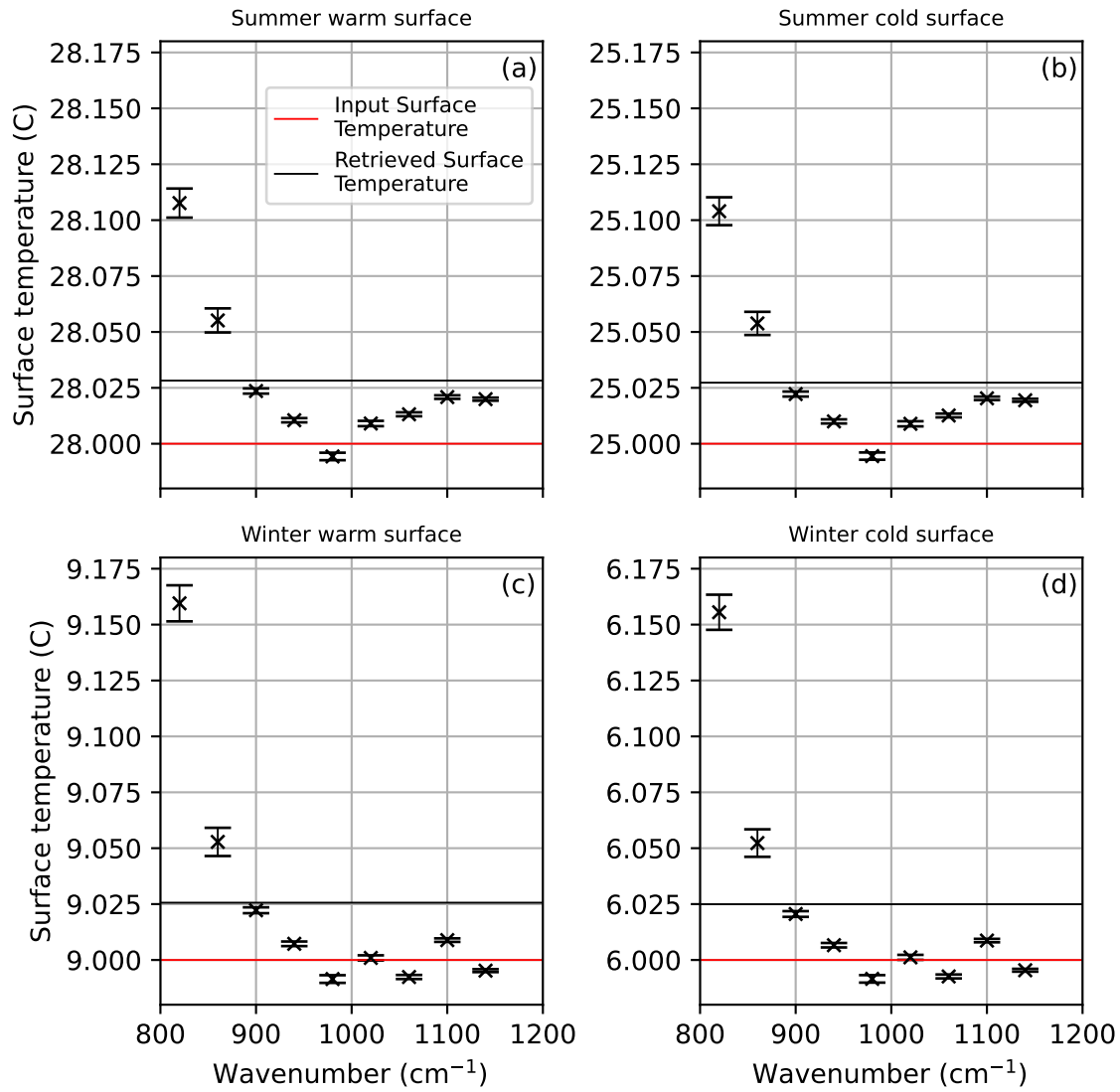


Figure 5.14: The retrieved surface temperature for each 40 cm^{-1} wavenumber bin between 800 and 1200 cm^{-1} , the wavenumber range chosen for the final retrieval. The red horizontal line is the input temperature and the black horizontal line is the average of the retrieved temperature in each bin. All y axes cover the same range.

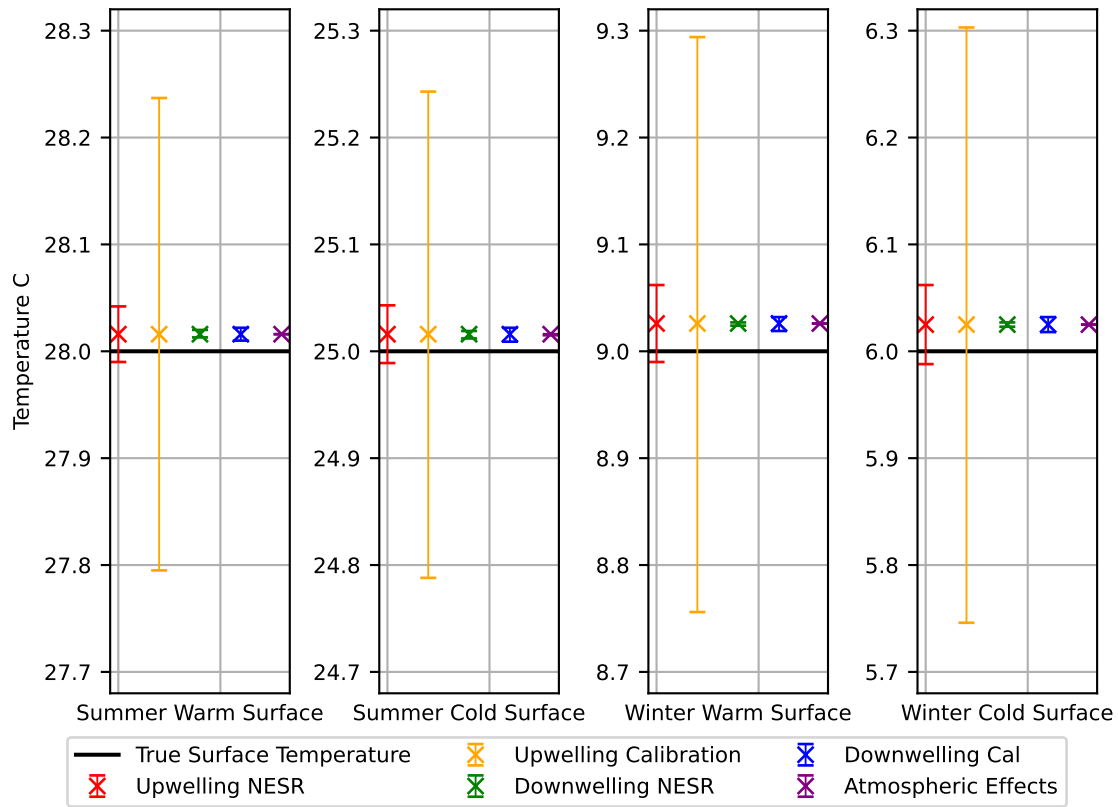


Figure 5.15: The retrieved surface temperatures for the four cases. The black line is the surface temperature specified in the simulation, the retrieval is plotted five times for each case with the error bars representing the effects of the perturbations listed in the legend.

larger in the winter case and the effects of the perturbations are larger, despite the perturbations being a similar size between the winter and summer cases. This suggests that the surface temperature retrieval is more sensitive to the input spectra in the winter case, possibly due to the lower surface temperature and hence lower radiance emitted from the surface. The perturbation that causes the largest change in the surface temperature is the calibration uncertainty in the upwelling spectrum. The changes to the downwelling spectrum and atmospheric transmission have a negligible effect on the retrieved surface temperature. Further work should be undertaken to investigate whether the bias in the temperature retrieval tends to be positive in a wider range of cases.

5.4.4 Emissivity Retrieval

Figure 5.16 shows the emissivity retrievals for the four cases with no perturbations applied.

The most striking difference is seen between the retrievals undertaken with the warm surface

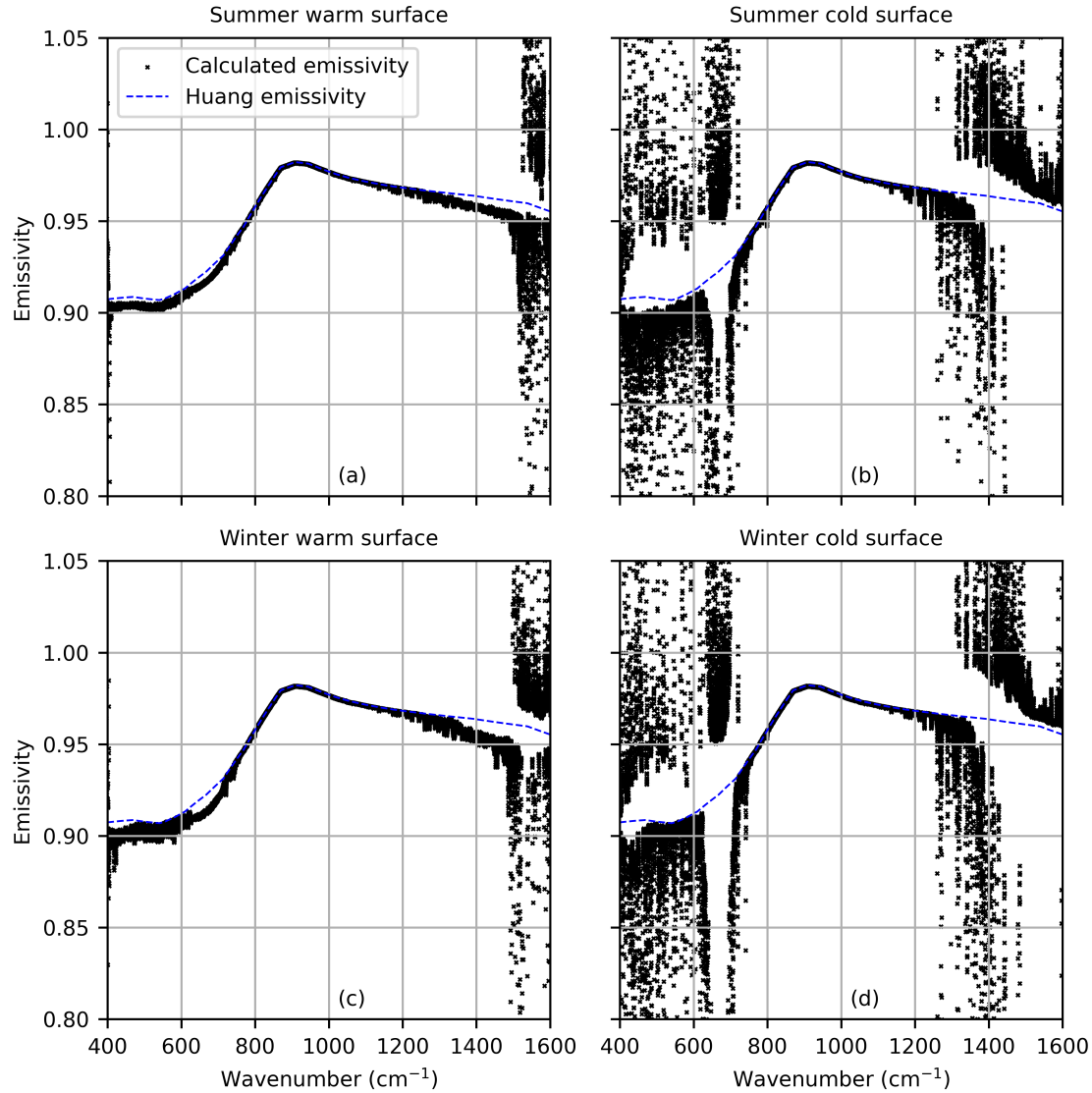


Figure 5.16: The emissivity retrieved using the FINESSE simulator for four different cases. The dashed line in each plot shows the input emissivity from the Huang et al. (2016) emissivity dataset.

and the retrievals undertaken with the cold surface. In the case of the cold surface, below 700 cm⁻¹ and above 1300 cm⁻¹ the retrievals split into two sets of values, with the lower set being a better match for the input emissivity. This splitting can also be seen above 1500 cm⁻¹

in the warm surface cases. The scatter of points in these regions is also larger than in the atmospheric window.

By examining the equivalent retrievals performed using the input surface temperature, figure 5.17, we can see that these differences between the input and retrieved surface emissivity are caused by the 0.025°C overestimation of surface temperature by the Newman method.

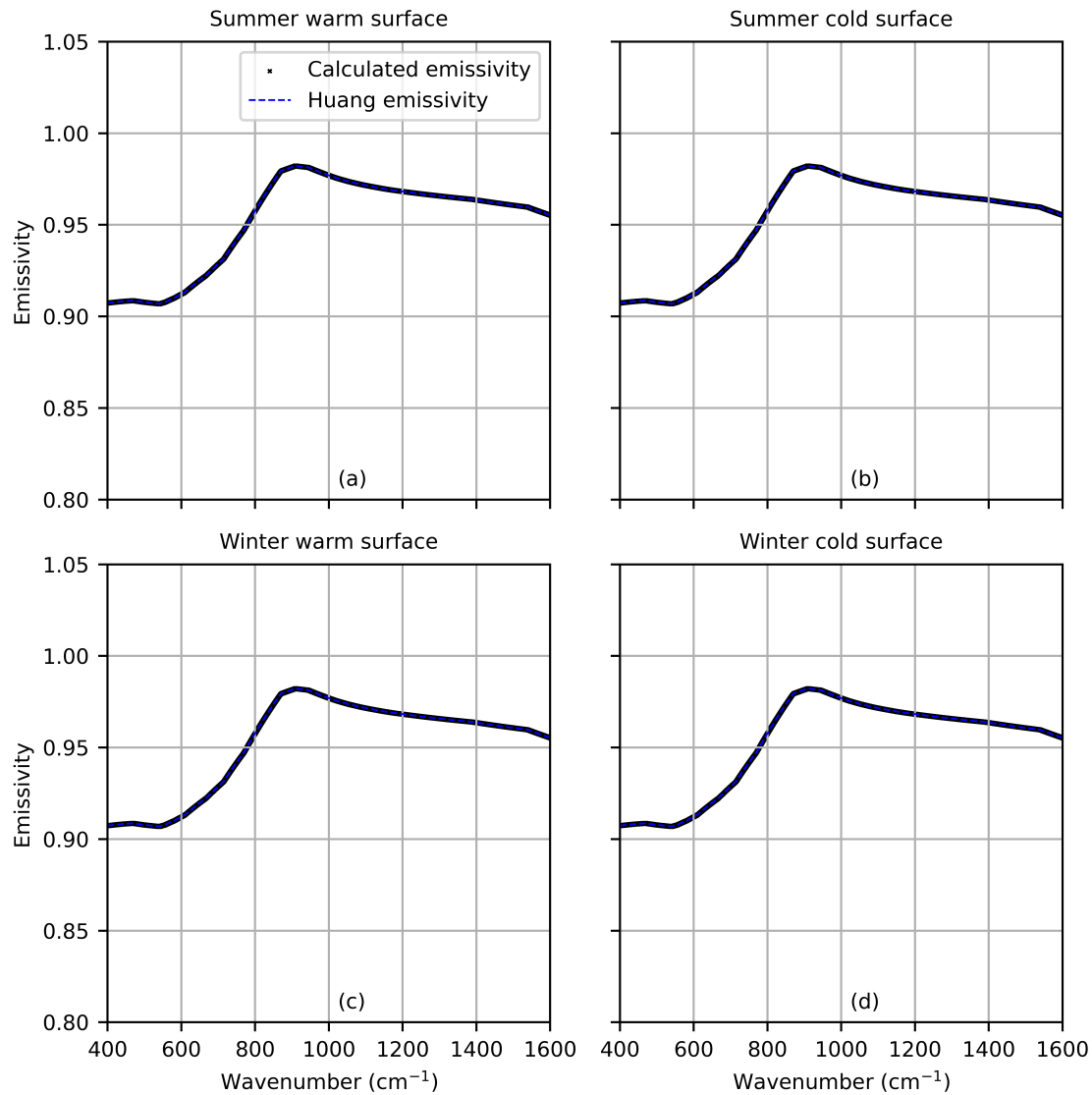


Figure 5.17: The same emissivity retrievals as figure 5.16 but using the input surface temperature rather than the temperature derived using the Newman method.

This behaviour of the emissivity retrieval, particularly the difference in behaviour of the warm and cold surface cases, can be more precisely explained by examining the derivative of equa-

tion 5.13 with respect to surface temperature:

$$\frac{d\epsilon}{dT_s} = \frac{-\epsilon}{B(T_s) - \tau L^\downarrow - (1 - \tau)B(T_a)} \frac{dB(T_s)}{dT_s} \quad (5.17)$$

$\frac{dB(T_s)}{dT_s}$ is the derivative of the Planck function with respect to temperature and is always positive as are the emissivity, transmission and radiance values. Therefore the behaviour of this equation depends on the sign of the denominator. This falls into three regimes depending on the relative sizes of $B(T_s)$ and $\tau L^\downarrow + (1 - \tau)B(T_a)$.

In cases where $B(T_s)$ is larger than $\tau L^\downarrow + (1 - \tau)B(T_a)$, i.e. that surface is comparatively warm compared to the overlying atmosphere, the negative sign in front of the equation indicates that in general the emissivity will decrease if the surface temperature increases, which is in line with intuition. The smaller the difference between the two terms, the more sensitive the retrieved emissivity value is to changes in the surface temperature. In the case of the warm surface retrievals in figure 5.16 the higher surface temperature retrieved using the Newman method has led to the retrieved emissivity being lower than the input emissivity across wavenumbers below 1500 cm^{-1} . Somewhat counter intuitively, this underestimation is smaller in the window region, where it is around 0.1%, and larger below 700 cm^{-1} and above 1300 cm^{-1} where it reaches up to 1% of the emissivity value.

In cases where $\tau L^\downarrow + (1 - \tau)B(T_a)$ is larger than $B(T_s)$ equation 5.17 is in the second regime where the retrieved emissivity increases as the surface temperature increases. Again the smaller the difference between the terms, the more sensitive the retrieved emissivity is to changes in surface temperature. This regime can be seen above 1500 cm^{-1} in the warm surface cases and below 700 cm^{-1} and above 1300 cm^{-1} in the cold surface cases. It occurs when the overlying atmosphere is more opaque and the downwelling radiance is high compared to the

direct radiance from the surface. The emissivity retrieval in these regions is also very sensitive to changes in the surface temperature.

Finally when $B(T_s)$ and $\tau L^\downarrow + (1 - \tau)B(T_a)$ are equal the behaviour is undefined, indicating that it is not possible to retrieve the surface emissivity when there is no contrast between the radiance from the surface and the downwelling radiance.

Figure 5.18 shows the effect that the perturbations listed in table 5.2 have on the retrieved emissivity in the summer warm surface case. The change in emissivity caused by the 0.025°C overestimation of surface temperature is also shown. The effect of every perturbation is largest below 800 cm^{-1} and above 1200 cm^{-1} . The largest individual contribution in the FIR is from the effect of the NESR on the upwelling spectrum, which reaches a value of 0.1 at 450 cm^{-1} and increases further towards 400 cm^{-1} . This is despite the effect of the calibration uncertainty in the upwelling spectrum causing the largest change in the retrieved surface temperature. Increasing the value of the measured upwelling spectrum and increasing the value of the measured transmission increases the value of the retrieved emissivity as expected given that the surface temperature is fixed. Conversely increasing the value of the measured downwelling spectrum decreases the retrieved emissivity.

Figure 5.19 is the same as figure 5.18 but for the summer cold surface case. The range of the y-axis has also been increased by a factor of 6. The large emissivity differences seen below 700 cm^{-1} and above 1300 cm^{-1} demonstrate how sensitive the retrieved emissivity is to perturbations in the input quantities. This is a consequence of the low contrast between the temperature of the surface and the temperature of the overlying atmosphere as discussed above.

The upper and lower bounds for the emissivity retrieval were then calculated for each of the

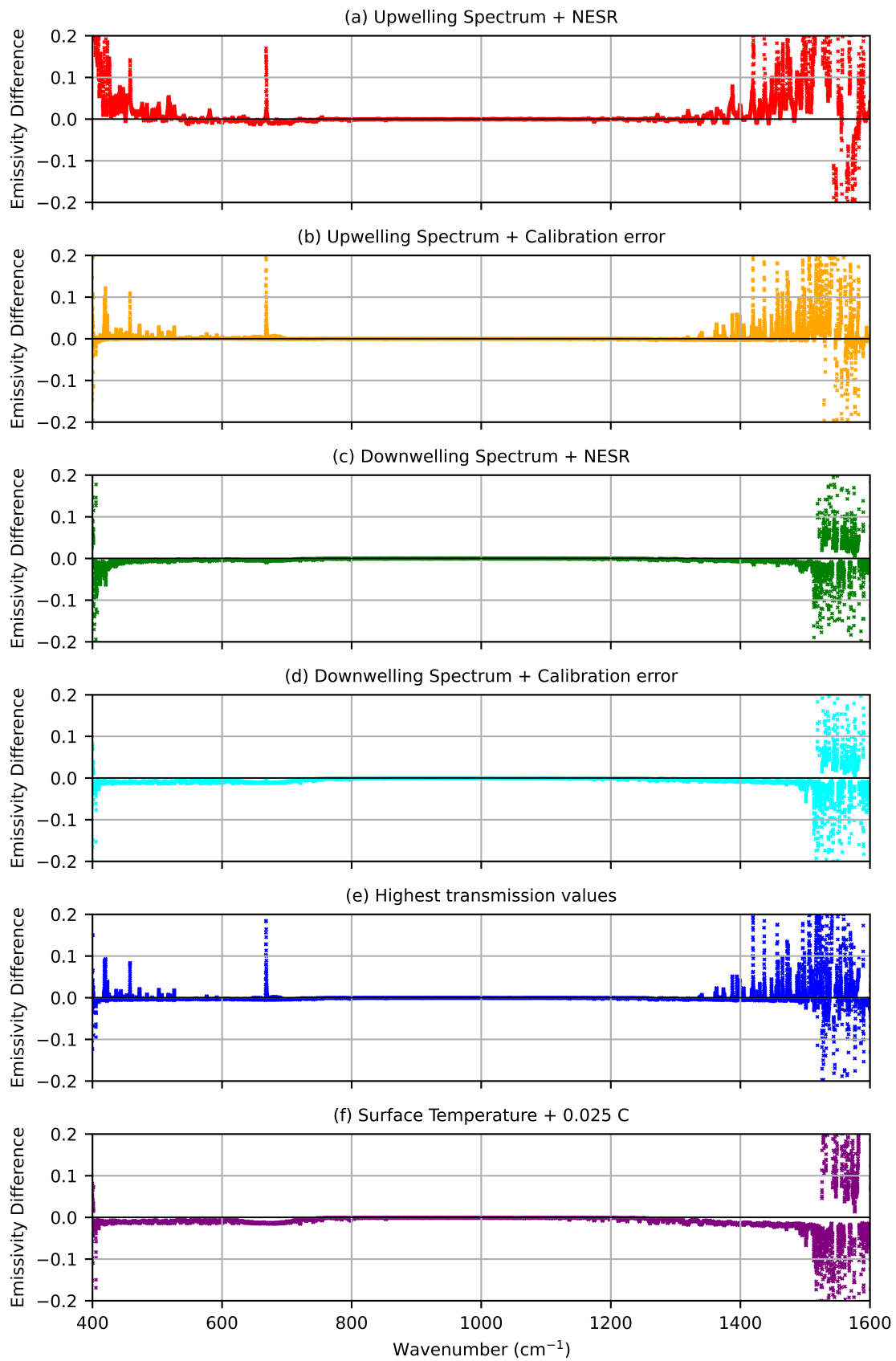


Figure 5.18: The change in emissivity caused by the labelled change in each parameter. All values are for the summer warm surface case.

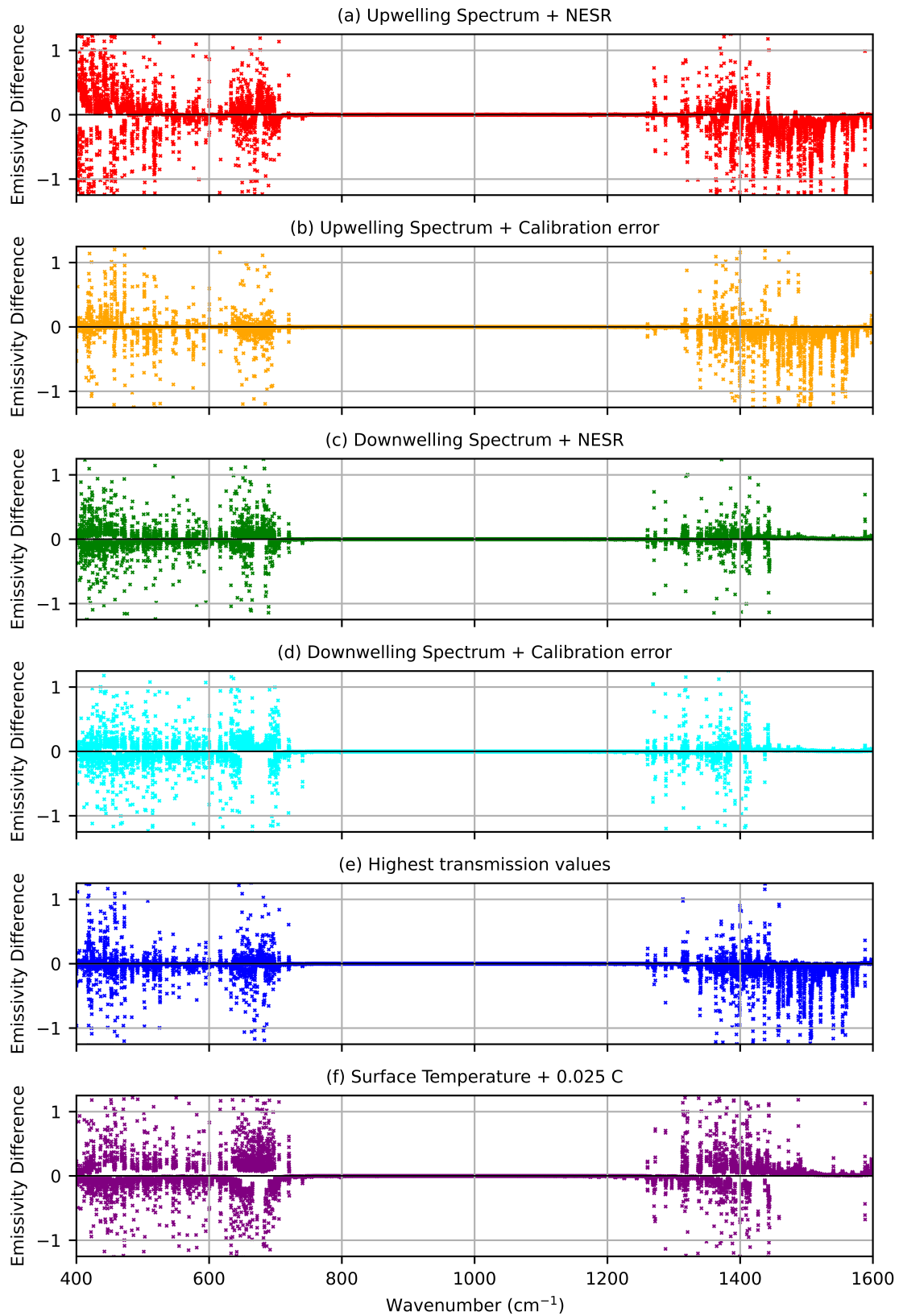


Figure 5.19: The change in emissivity caused by the labelled change in each parameter. All values are for the summer cold surface case.

four cases. For the upper bound this was done by adding in quadrature the effects of increasing the value of the measured upwelling spectrum by the NESR and the calibration uncertainty, increasing the value of the transmission spectrum, decreasing the value of the downwelling spectrum by the NESR and calibration uncertainty and decreasing the surface temperature. The opposite was done to calculate the lower bound of the emissivity. The percentage difference of the retrieved emissivity from the input emissivity is shown in figure 5.20 and the upper and lower bounds are shown by the grey shading. In all cases the change in emissivity caused by

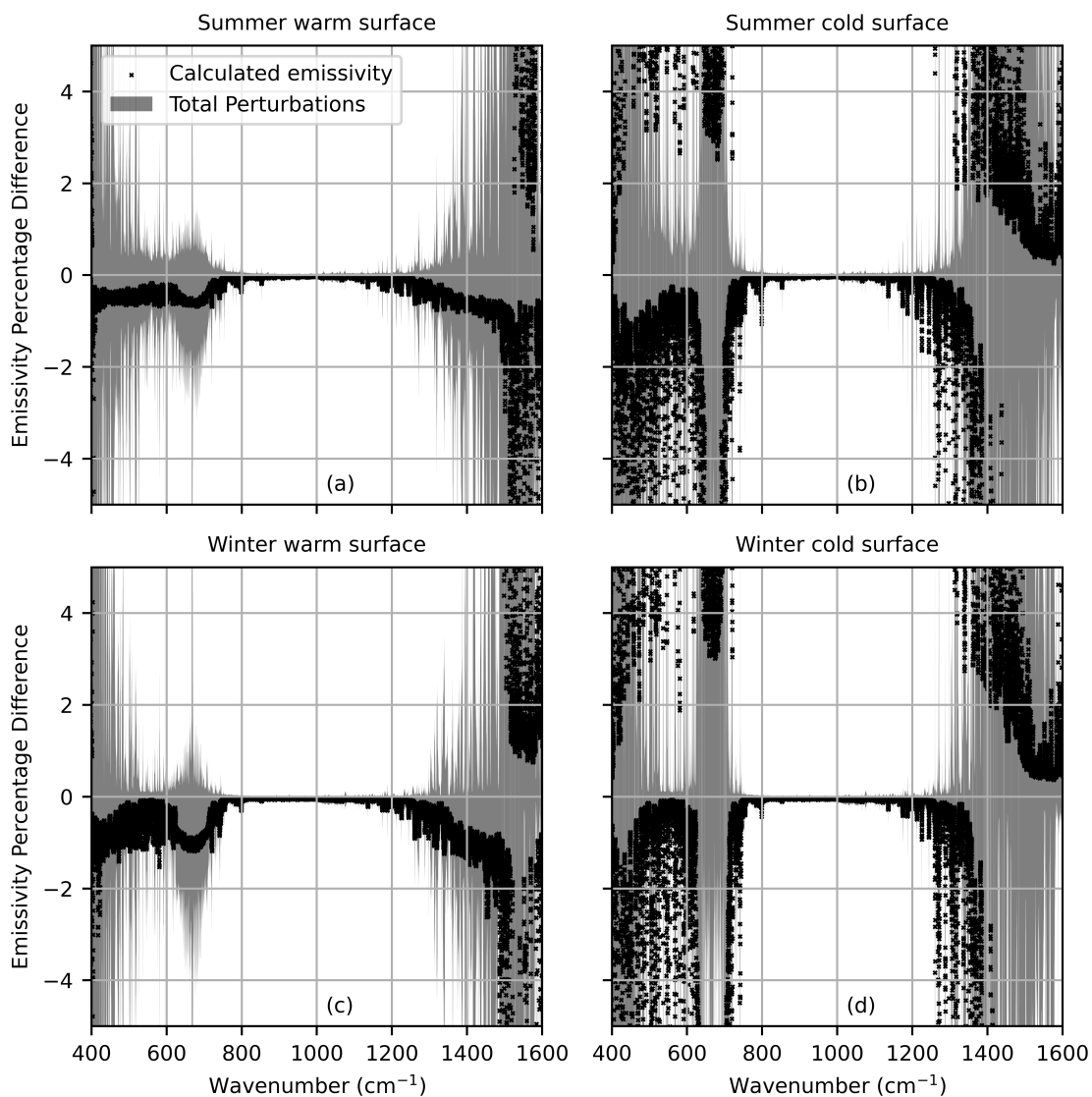


Figure 5.20: The percentage difference between the retrieved emissivity and the input emissivity for the four cases. The grey shading represents the upper and lower bounds of the retrieved emissivity when the perturbations are applied.

the combination of all perturbations is around $\pm 0.05\%$ between 800 and 1200 cm^{-1} . This increases to $\pm 1\%$ between 480 and 650 cm^{-1} in the warm surface cases and increases further below 480 cm^{-1} . The total uncertainty is larger in the cold surface cases.

Overall the real emissivity retrievals from FINESSE can be expected to have smaller uncertainties in the MIR window compared to the FIR and at wavenumbers in excess of 1200 cm^{-1} . This is to be expected given the enhanced transmission in the window region and high contrast between the surface emission and downwelling radiation at the surface. Given the expected FINESSE performance the emissivity can be retrieved in the range 800-1200 cm^{-1} with an uncertainty of $\pm 0.05\%$. In the cases where the surface temperature is heated to 2 K above the ambient air temperature the emissivity can be retrieved in the range 400-600 cm^{-1} with an uncertainty of $\pm 1\%$. In the cases where the emitting surface is 2 K colder than the ambient air temperature, there are still regions in the 400-600 cm^{-1} range where the emissivity can be retrieved to within 2% of the input value. This suggests that with appropriate filtering of the data, surface emissivity can be retrieved to this accuracy in these conditions across the 400 to 600 cm^{-1} spectral range. This is of particular interest for retrieving emissivity over snow and ice in the field. The retrievals from snow and ice in the field correspond most closely to the winter cold surface scenario due to typically cold atmospheres and the occurrence of temperature inversions where the surface is cooler than the overlying atmosphere (e.g. Zhang et al., 2011).

5.4.5 Data Filtering

The aim of this section is to investigate the performance of possible filters for the retrieved emissivity which will preferentially remove data in regions where the emissivity retrieval is too

sensitive to be of practical use while retaining as much data as possible.

Two criteria will be used to assess the performance of these filters. Firstly, the RMS difference between the input emissivity and the retrieved emissivity. A successful filter should reduce this value as it should remove data points that are far from the input emissivity while retaining data points that are close to the input emissivity. The second criterion is the fraction of points remaining, as a method that removes fewer points to achieve the same reduction in RMS difference is more specifically targeted at the regions where the retrieval is difficult.

Two methods of data filtering will be considered. Both filtering methods are applied to the final emissivity retrieval after all calculations including surface temperature retrieval have been performed. The first method removes the emissivity retrieval where the transmission between the surface and detector is below a specified value. This method was used by Bellisario et al. (2017) when retrieving snow and ice emissivity from a low-flying aircraft.

The second method is based on the work done in section 5.4.4 which showed that the emissivity retrieval becomes very sensitive to perturbations in the surface temperature when $B(T_s)$ and $\tau L^\downarrow + (1 - \tau)B(T_a)$ are similar in size. The quantity $B(T_s) - \tau L^\downarrow - (1 - \tau)B(T_a)$ also appears directly in equation 5.13 and in the derivative of equation 5.13 with respect to the upwelling spectrum (see appendix A). This method removes data when the difference between these values is below a specified threshold and will be referred to as the contrast filter.

Figure 5.21 shows the effect of the transmission filter for transmission cutoff values of 0.95, 0.99 and 0.999. As the transmission threshold is increased from 0.99 to 0.999, more points are excluded, with those remaining tending to be closer to the input emissivity. This can be seen in the reducing RMS values for the winter cold surface case, which are shown in table 5.3. However a transmission limit of 0.95 increases the RMS difference as some points closer to the

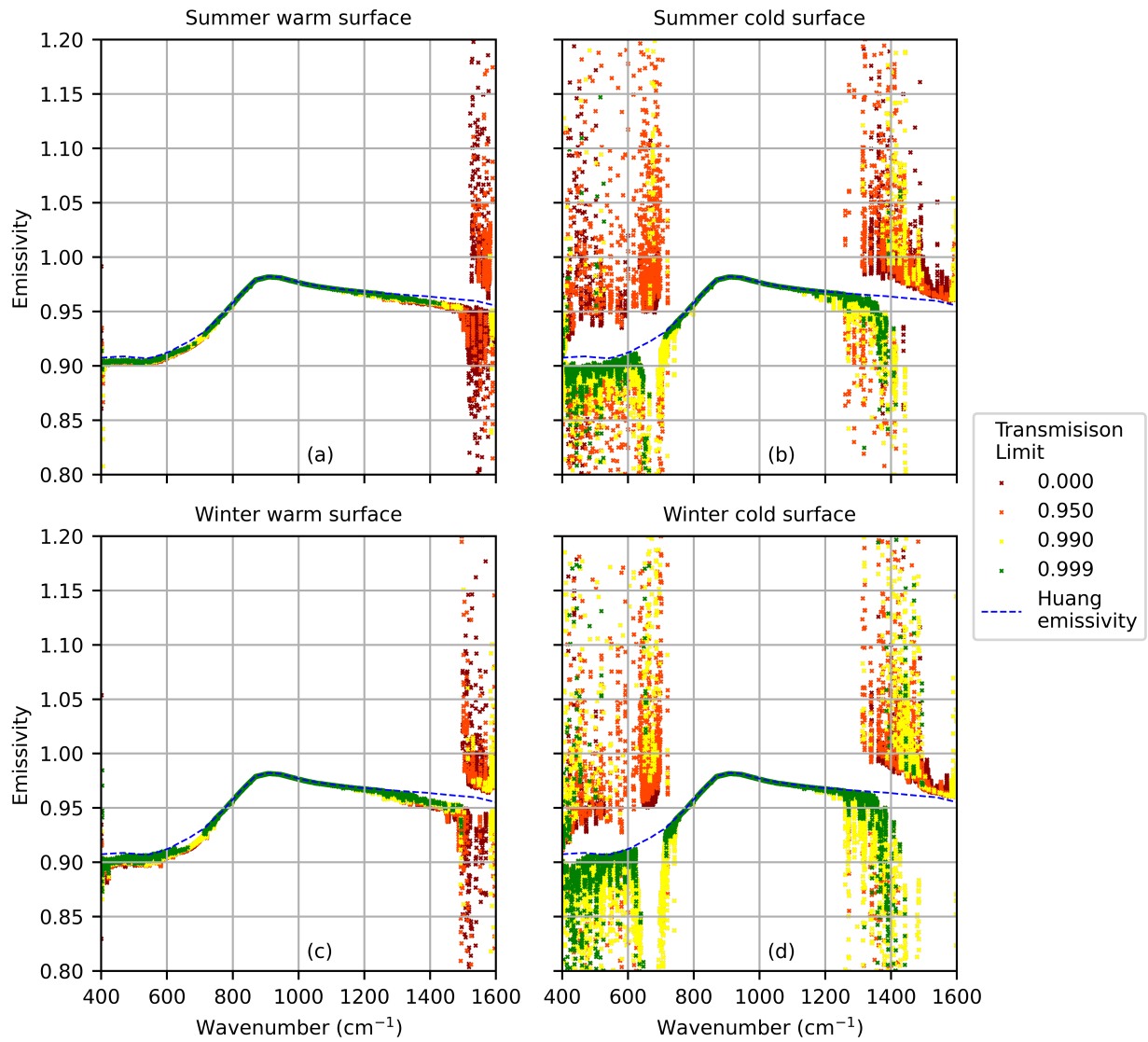


Figure 5.21: The emissivity retrieval with the transmission filter applied at three different cutoff values.

input emissivity are removed while points further from the input emissivity remain. Even at a transmission limit of 0.999, which removes on average 45% of the points across all four cases, some anomalous values are still present in the cold surface cases.

Method	RMS difference	Fraction of points remaining
None	2.366	1
Transmission		
0.95	2.478	0.90
0.99	1.175	0.78
0.999	0.076	0.58
Contrast		
5×10^{-4}	0.015	0.82
1×10^{-3}	0.006	0.71
5×10^{-3}	0.001	0.53

Table 5.3: The RMS difference from the input emissivity and the fraction of points remaining for different filtering methods. These data are from the winter cold surface spectrum.

Figure 5.22 shows the effect of the contrast filter for different values of contrast. This technique can very specifically remove points based on how far they are from the input emissivity. This allows the RMS to be reduced while retaining a higher proportion of the data points, as seen in table 5.3. This specificity is somewhat expected as in this case any deviation from the input emissivity is only caused by the incorrect surface temperature.

Applying the contrast filter with a contrast level of $0.005 \text{ W m}^{-2} \text{ sr}^{-1} \text{ cm}$ to the data in figure 5.20, selects data with a total error of less than $\pm 2\%$ below 600 cm^{-1} in the cold surface cases. This is plotted in figure 5.23. The total error in the window region is unchanged as the data is retained in this region.

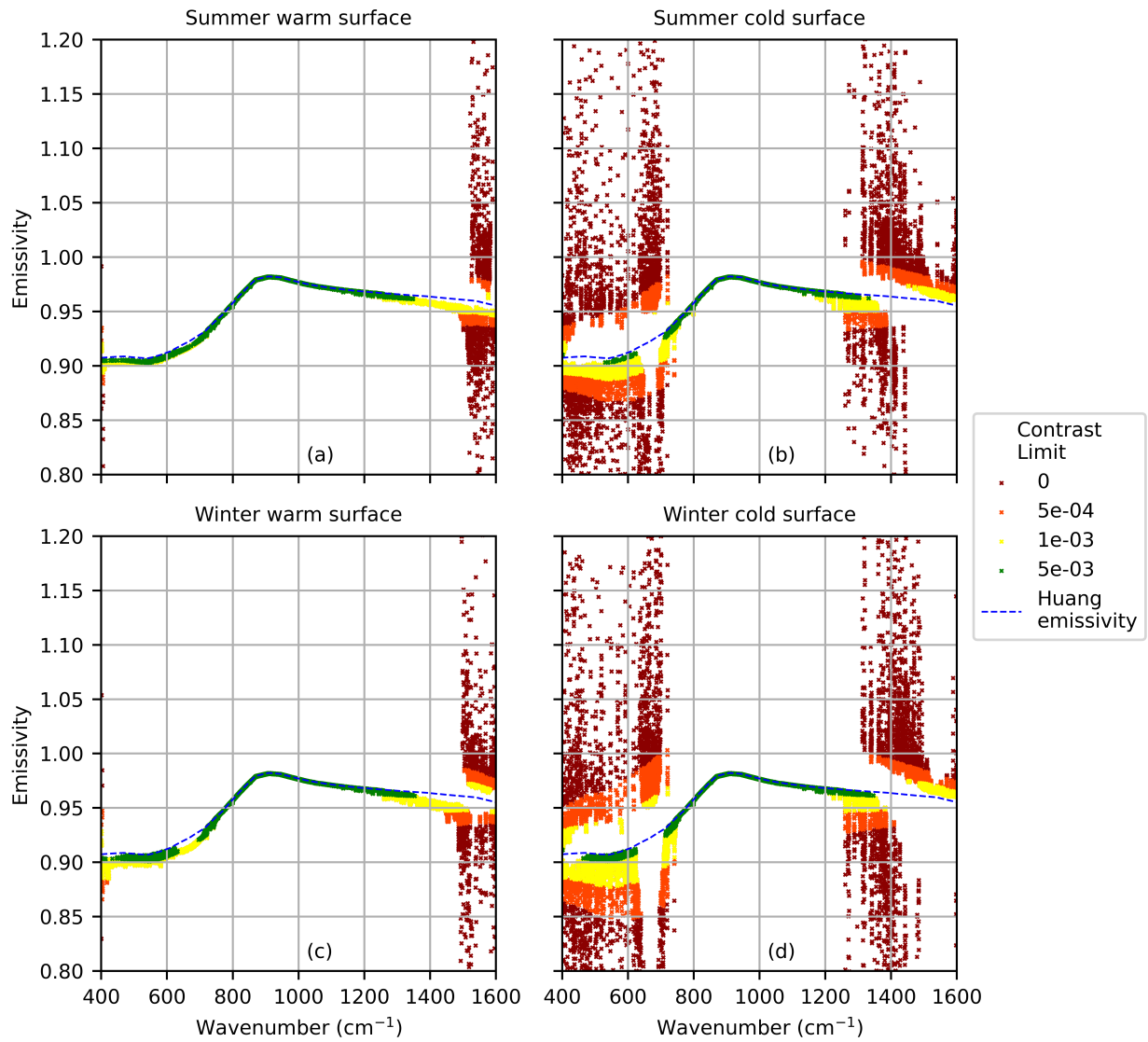


Figure 5.22: The emissivity retrieval with the contrast filter applied at three difference values.

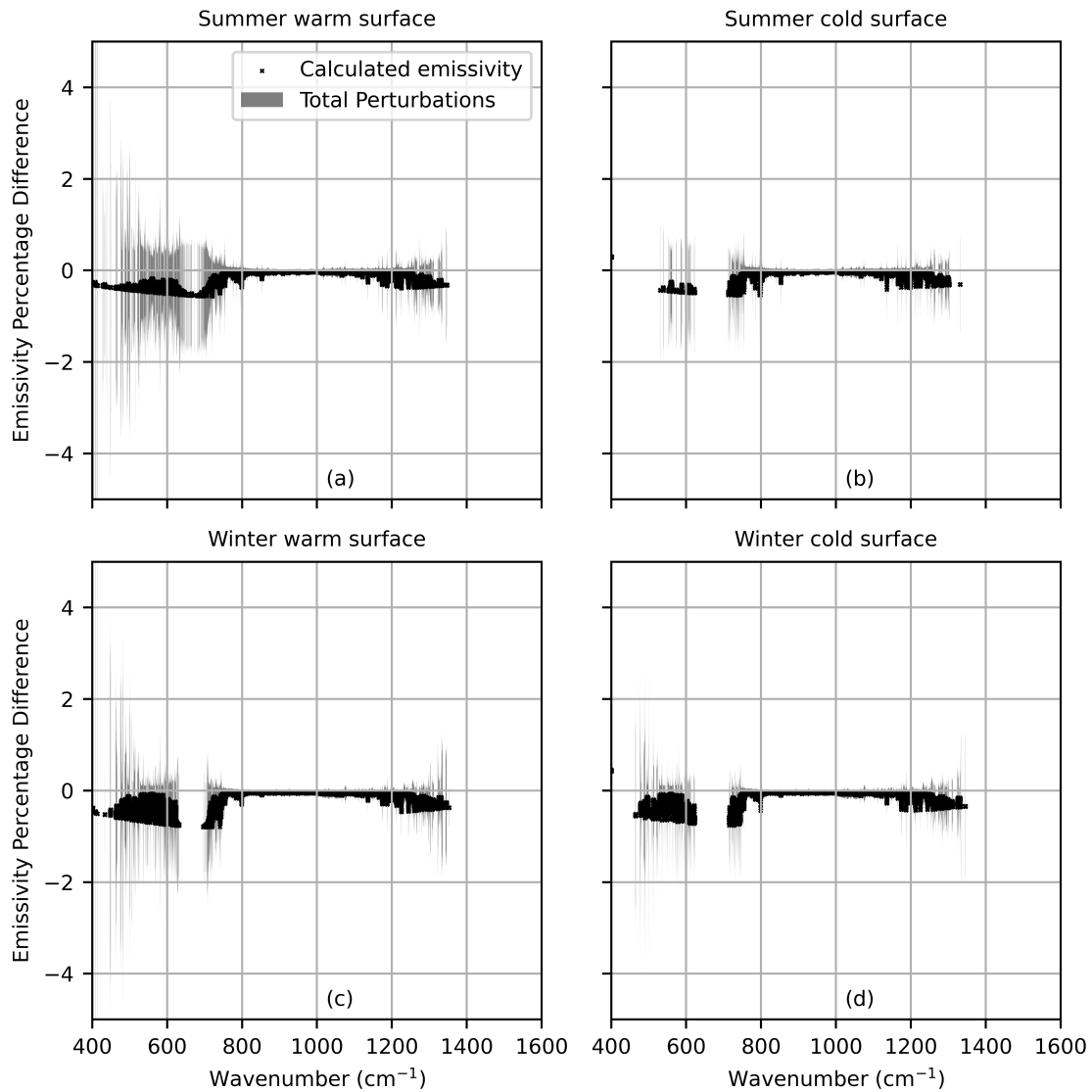


Figure 5.23: The percentage difference between the retrieved emissivity and the input emissivity for the four cases as in figure 5.20. The input emissivity is taken from the Huang et al. (2016) dataset. The data have been filtered using the contrast filter set at a level of $0.005 \text{ W m}^{-2} \text{ sr}^{-1} \text{ cm}$.

5.5 Conclusion

This chapter has introduced the FINESSE instrument along with the calibration process, the method for determining the measurement uncertainty, and the instrument line shape. The chosen emissivity retrieval method was explained. The FINESSE simulator was also developed and used to explore the performance of retrievals of water emissivity in the atmospheric conditions seen in London during the summer and winter.

The work with the FINESSE simulator demonstrates that surface emissivity retrieval in the FIR under the conditions expected in London is possible using FINESSE. However below 700 cm^{-1} the emissivity retrieval is much more sensitive to changes in the input quantities than it is in the MIR window. For the conditions investigated in this chapter, the Newman temperature retrieval method could successfully retrieve the surface temperature to within 0.025°C , with the retrieved temperature higher than the input surface temperature. It was demonstrated that the retrieved surface emissivity is highly sensitive to the surface temperature, particularly in the FIR and when there is low contrast between the temperature of the surface and the temperature of the overlying atmosphere. A key area of future work is the refinement of the temperature retrieval, possibly by increasing the range over which the surface temperature is determined and undertaking further tests to determine whether the positive bias seen in these two cases is a more general rule. Another option is to investigate other methods that rely on spectral smoothness such as that used by J. E. Murray et al. (2020) or use a different method that assumes a transmission of one (Salvaggio & Miller, 2001), but only in the wavenumber range where this value is appropriate.

In conditions where the surface was warmer than the ambient air, the largest change in the retrieved emissivity was caused by increasing the upwelling spectrum by the value of the NESR

and the total change in the emissivity caused by the combination of all the perturbations considered was $\pm 1.5\%$ between 450 and 600 cm^{-1} . The retrieved emissivity was much more sensitive to changes in the input conditions when the surface was cooler than the ambient air temperature, however by filtering the data it was possible to isolate regions between 400 and 600 cm^{-1} where the emissivity could be retrieved. In these regions, the total change in the emissivity caused by the combination of all the perturbations considered was around $\pm 2\%$. The contrast filter developed in this chapter will be used in chapter 6. However in the future, further ways of filtering the data should be explored.

The simulator is a valuable tool to investigate the potential performance of FINESSE under different atmospheric conditions. This is particularly useful as FINESSE will be deployed in the field, possibly in arctic environments. The simulator will allow us to test the retrieval before FINESSE is deployed and make changes to the measurement method or analysis techniques. For example the performance of emissivity retrievals carried out overnight compared to during the day could be explored. Furthermore the timing of fieldwork could be decided based on the climatological conditions that would result in the best measurements. The simulator could also be used to investigate more unusual experimental setups, for example the quality of the retrieval is dependent on the contrast between the upwelling and downwelling radiance, so the simulator could be used to explore the possibility of achieving this contrast using a radiator above the surface.

The development and characterisation of FINESSE described in this chapter will enable us to carry out the emissivity retrievals in chapter 6. The tests with the FINESSE simulator will also aid the interpretation of the resulting retrievals.

Chapter 6

Measurement of Water Emissivity

This chapter presents measurements of the emissivity of deionised water in the far- and mid-infrared made using FINESSE. The choice of deionised water means it is possible to compare the measurements to previous measurements made in the MIR and to theoretical calculations. This will enable an assessment of the performance of FINESSE and the retrieval method that has been developed. Section 6.1 outlines the experimental setup and the measurements undertaken. In section 6.2, the response function is calculated and its stability is assessed. Section 6.3 describes the method for calculating the emissivity. Emissivity retrievals for several cases covering a variety of atmospheric and surface conditions are shown and discussed in section 6.4. In section 6.5 these measurements are compared to theoretical calculations and in sections 6.6 and 6.7 the dependence of the emissivity on surface temperature and viewing angle is explored. To our knowledge, at the time these measurements were made there were no observationally based estimates of the emissivity of water in the FIR.

6.1 Measurements Undertaken

In this chapter I will discuss five sets of water emissivity measurements made using FINESSE. These were taken from the rooftop of Imperial College London under clear sky conditions on 25-Feb-19, 08-Sep-21, 11-Feb-22 and 17-Mar-22 and are summarised in table 6.1. The 25-Feb-19 measurements were taken by MSci students Micol Bedarida and Rachel Sfeir assisted by Jon Murray and me. I took the remaining measurements with assistance and advice from Jon Murray.

Measurement Set	Test	1	2	3	4
Date	25-Feb-19	08-Sep-21	08-Sep-21	11-Feb-22	17-Mar-22
Time	1115-1715	0938-1100	1130-1252	0927-1237	0735-1257
Duration	6h00	1h22	1h22	3h04	5h22
Water Temperature (°C)	17 (ambient) and 35 (cooling slowly)	20 (ambient)	30 (heated)	22-14 (cooling slowly)	8-15 (ambient)
View Angle	55	50	50	45	various
Atmospheric Temperature (°C)	14-18	22-24	26-30	6-11	5-14
Humidity (%RH)	50-30	65-55	50-40	70-40	70-40
Path Length (cm)	162	34	34	32	various

Table 6.1: Measurements undertaken using FINESSE which are discussed in this thesis.

6.1.1 Test Measurements

These measurements were taken while FINESSE was still under development. Specifically, the optical window was 10 μm thick polypropylene as the diamond window had not been installed and the additional environmental monitoring equipment was not present. Before

these measurements took place the accuracy of the FINESSE viewing angle was confirmed in the laboratory. Reference points were marked on the floor at fixed angles from the FINESSE calibration system. The spectrometer was then removed from the calibration system and a laser was shone through the calibration system. The calibration system was then directed to view the reference points and the stepper motor controlling the pointing mirror was adjusted until the laser lined up with the references.

The test measurements were undertaken under completely clear sky conditions. Humidity was generally low at around 40% and the air temperature was between 14 and 18°C. FINESSE undertook radiance measurements of a water surface held in a roughly 40x60 cm tray and of the clear sky. These measurements are discussed in section 6.4.

6.1.2 1st and 2nd Measurement Sets

These measurements were taken on a hot, sunny day in early September 2021. The external air temperature reached 30°C by 1200 BST, this caused the ambient blackbody to gradually increase in temperature throughout the day. Measurements were made of deionised water, this was initially at ambient temperature and was then heated using a Grant Instruments T100 water bath to a temperature of around 30°C. The water bath had a width of 23 cm and a length of 37 cm. The diameter of the FINESSE field of view was calculated to be 12 cm at the surface of the water. Therefore we were confident that FINESSE was only viewing the surface of the water. Both the 1st and 2nd sets of measurements are discussed in sections 6.4 and 6.5. The 2nd set of measurements are also used in section 6.6 to investigate the temperature dependence of the emissivity.

6.1.3 3rd Measurement Set

This set of measurements was undertaken in cold, clear conditions in February 2022. The water was initially heated to 30°C using the water bath but this produced a noticeable amount of mist above the water bath. Therefore, the water was allowed to cool, and measurements were started once the mist had dissipated. Figure 6.1 shows a photograph of the setup for the emissivity measurements of heated water. When water view measurements were underway the water bath circulator was switched off to avoid the rippling of the water surface that is visible in this photograph. It was important to ensure the surface of the water was as flat as possible as the analysis and subsequent comparison to theory assumed that the surface of the water acted as a flat specular reflector. These measurements are discussed in sections 6.4 and 6.5.



Figure 6.1: FINESSE undertaking measurements of heated water during the 3rd set of emissivity measurements.

6.1.4 4th Measurement Set

These measurements were undertaken in very similar atmospheric conditions to those described in the previous subsection. The aim of these measurements was to determine the emissivity of water at a variety viewing of angles. The water was held in a long tray, allowing measurements to be made at angles of 50, 60 and 70°. Figure 6.2 shows the setup for these angular measurements. The screen was required as a shelter from the wind because, although the wind was very light, the geometry of the tray made the water very susceptible to ripples. The 60° measurements are used in sections 6.4 and 6.5. The 50° measurements are used in section 6.6 to investigate the temperature dependence. The measurements at all three angles are used in section 6.7 to investigate the angular dependence of the emissivity.



Figure 6.2: Photograph of the setup for FINESSSE to make measurements of water emissivity at different angles.

6.2 Determining the Response Function

Determining the emissivity measurement cycles is the first step to retrieving emissivity. Each emissivity measurement cycle consists of a surface view at angle θ accompanied by calibration views of the hot and ambient black bodies, and a sky view at an angle of $180 - \theta$ with corresponding calibration views. Each scene view is paired with the sky view which is closest in time, therefore the order of scene and sky views is not fixed, and a single sky view can be included in up to three cycles.

The next step is to calculate the response function for each hot black body, ambient black body, scene triplet. This is done using the method described in section 5.1.2. The average response function for each set of measurements is shown in figure 6.3a and the uncertainty in the response function due to uncertainty in the temperatures of the HBB and ABB is shown in figure 6.3b.

The response function for the test measurements is higher due to the higher transmission of the thin polypropylene window compared to the diamond. However polypropylene absorption features are visible (e.g. 820 cm^{-1} , 1000 cm^{-1} , 1150 cm^{-1}). The response function for the diamond window is lower but smoother, without the sharp absorption features. The response functions for measurements 3 and 4 taken in winter 2022 are slightly lower across the whole spectrum. This may be due to the different environmental conditions between the two sets of measurements or the slight degradation of the optical components over time. All the response functions show a sensitivity to radiation above 400 cm^{-1} . Spectral features can also be seen in all the response functions. These include absorption by the CO_2 band centred at 667 cm^{-1} and by water vapour between 400 and 600 cm^{-1} and above 1300 cm^{-1} . These are caused by the presence of water vapour and CO_2 both in the path between the black body targets and

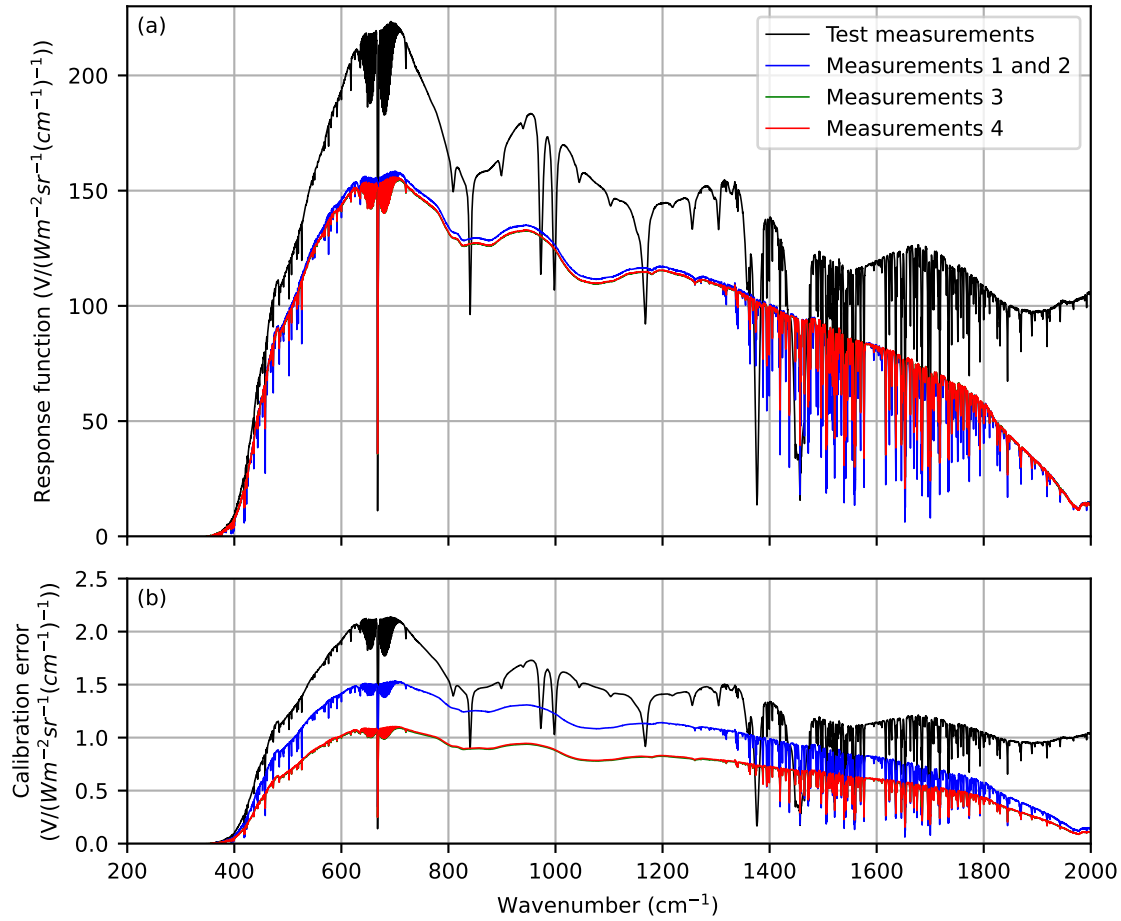


Figure 6.3: (a) Average response functions for the five sets of measurements discussed in this chapter. The response functions for measurements 3 and 4 are almost identical. The response function is larger for the test measurements due to the polypropylene window however it also contains noticeable polypropylene absorption features. (b) The uncertainty in the response function due to uncertainty in the temperatures of the HBB and ABB calibration targets.

the window and within the interferometer itself. The calibration error is around one percent of the measured spectrum for the first and second sets of measurements. It is lower for the third and fourth sets of measurements due to the larger temperature difference between the HBB and ABB during the winter measurements.

Examining the evolution of the response function over time allows us to investigate the stability of the instrument. This is done by calculating the percentage difference between a single response function and the average response function for the whole day's measurements. Figure 6.4 shows this percentage difference from the average response function plotted against time

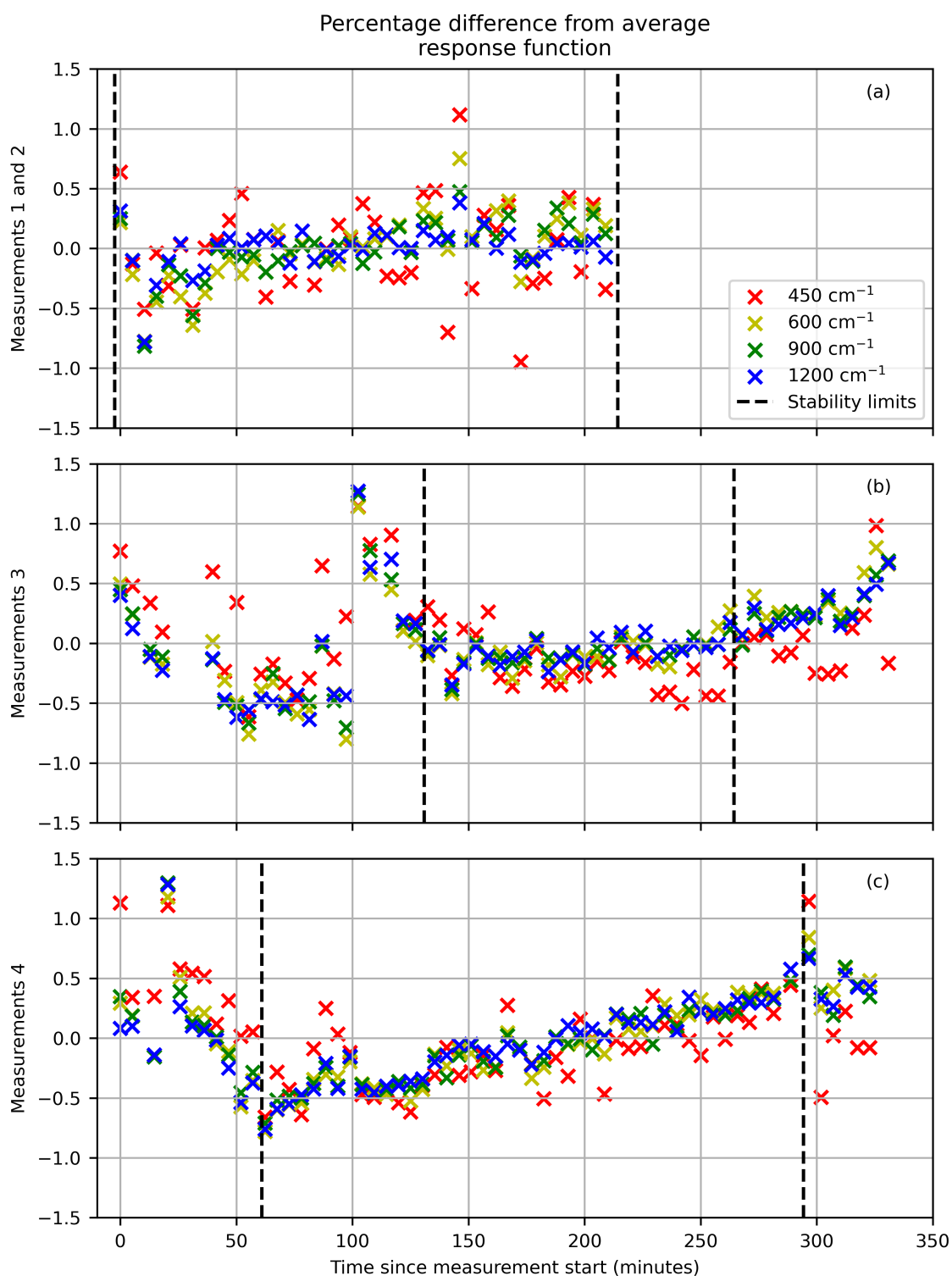


Figure 6.4: Percentage difference from the average response function against time for four wavenumber bands. The dashed black lines show the stability limits determined for each set of data.

for the four numbered sets of measurements. The four different colours represent 4 wavenumber bands each with a width of 1 cm^{-1} .

All three plots show variation in the response function between each calibration cycle and also longer timescale shifts in the response function that occur over the course of tens to hundreds of minutes. They also show the response function taking some time to stabilise. This is due to the time taken for the HBB target to reach the set temperature. The longer timescale shifts are not observed in data taken in the laboratory (Jon Murray, personal communication 2022) so they are likely caused by the gradually increasing temperatures throughout each measurement day. The short term variation occurs between each 5 minute calibration cycle and is around 0.5% once the system has stabilised. This is smaller than the uncertainty in the response function due to uncertainty in the temperatures of the calibration targets (figure 6.3b). This short term variation of the response function is slightly larger for the summer measurements (figure 6.4a). In earlier test measurements it was also seen that the response function changed quickly in response to the instrument coming under direct solar illumination. The short term variation in the response function is of particular interest as this may indicate changes in the response function of the instrument within a calibration cycle. Any change in the response function within the calibration cycle will impact the accuracy of the calibrated radiances. The slower shift of the response function does not impact the calibrated radiances as we use the closest response function in time to calibrate our measurements. However, in the future, a systematic study of the rate of change in the response function would allow us to minimise the time spent on calibration views without impacting the accuracy of the calibration.

For the measurements in this thesis, the emissivity was only retrieved in regions where the response function was stable. I defined this as regions where the response function was not changing rapidly or exhibiting discontinuities (such as at 100 minutes in figure 6.4b) and the

short term variation in the response function was less than roughly 0.5%. The stable regions were identified visually using figure 6.4 and are marked by the black dashed lines.

6.3 Calculating the Emissivity

Once the stable region and the emissivity cycles have been determined, the transmission between the surface and the instrument is calculated. This is done using LBLRTM for each emissivity cycle that falls within the stability limits. LBLRTM is run in horizontal mode at a resolution of 0.001 cm^{-1} using the distance between the surface and pointing mirror. The pressure, temperature, humidity, and CO_2 concentration are each determined by averaging the corresponding measurements from the Vaisala sensors over the course of the whole emissivity cycle. The atmosphere is assumed to be homogeneous. The transmission is then apodised using the ILS described in section 5.1.4. Figure 6.5 shows an example transmission spectrum before and after the apodisation.

Finally, the emissivity and surface temperature are jointly retrieved for each emissivity measurement cycle using the method described in section 5.2. The surface temperature was retrieved using the wavenumber range 800 to 1200 cm^{-1} with bins of 40 cm^{-1} .

6.4 Emissivity Retrievals

Figure 6.6 shows the retrieved emissivity for the test measurements undertaken when the polypropylene window was installed. This is the average of one hour of measurements taken when the water was heated.

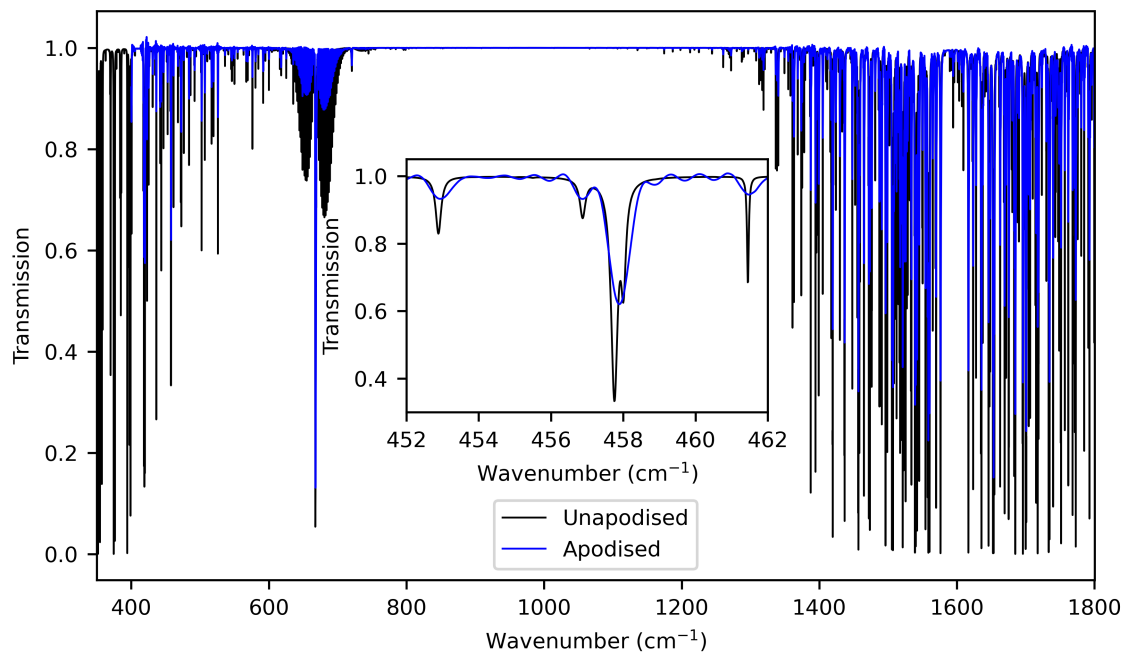


Figure 6.5: An example transmission simulation, before (black) and after (blue) the application of the ILS. Inset: detail of several spectral lines before and after apodisation.

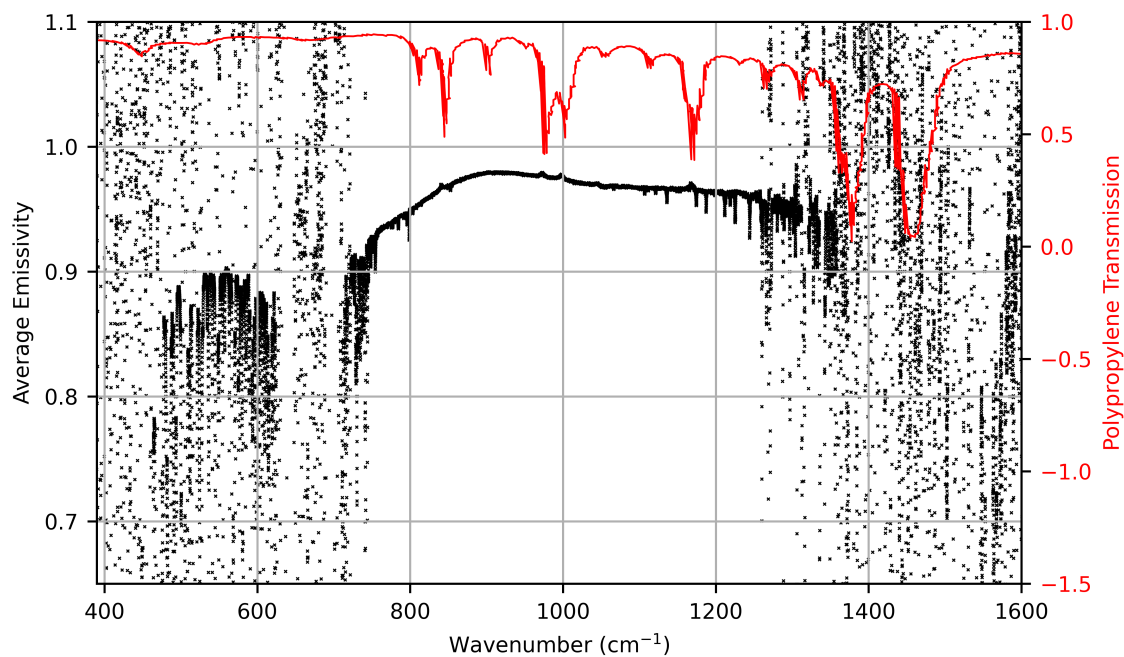


Figure 6.6: Emissivity from the test measurements (black) and the transmission spectrum of polypropylene (red).

In the MIR the retrieval accurately produces the expected emissivity for water (see figure 1.5) with a low scatter of points however below 700 cm^{-1} and above 1300 cm^{-1} the scatter in

the retrieved emissivity value increases dramatically and includes values that are not physical. There are also small spikes visible in the retrieval at 850, 970 and 1000 cm^{-1} . As the red line on figure 6.6 shows, these are regions of polypropylene absorption. These spikes were also visible in the calibrated spectra. These results provided the motivation to change the window material from polypropylene to diamond, which has a flatter transmission spectrum across the MIR and FIR, as evidenced in figure 6.3.

Figure 6.7 shows example water and sky view spectra used for the retrieval of water emissivity. The spectra are taken from measurement sets 1-4 and were chosen to illustrate a range of atmospheric conditions, surface temperatures and view angles. Each plot shows the average of 8 cycles corresponding to 1.5 hours of measurement time. The NESR and calibration error for each spectrum is also shown with the NESR reduced by the averaging of the spectra. As seen in figure 5.4 the calibration error is higher in the window region and is generally higher than the NESR.

Figure 6.8 shows the retrieved surface temperature in each 40 cm^{-1} wavenumber bin between 800 and 1200 cm^{-1} . The error bars show the uncertainty associated with the inversion of the Planck function. The horizontal line shows the average of the temperatures and this is taken to be the surface temperature for that spectrum. The scatter of the retrieved surface temperature in each bin is larger than that seen in the simulator (figure 5.14) and similar across all sets of data. This means that the retrieved surface temperatures in the 800 and 840 cm^{-1} bins are no longer clear outliers. This is likely due to the random noise on the measured spectra as this affects each wavenumber independently, which makes the measured spectra less smooth. Further work should be done to investigate the optimal wavenumber range for the surface temperature retrieval.

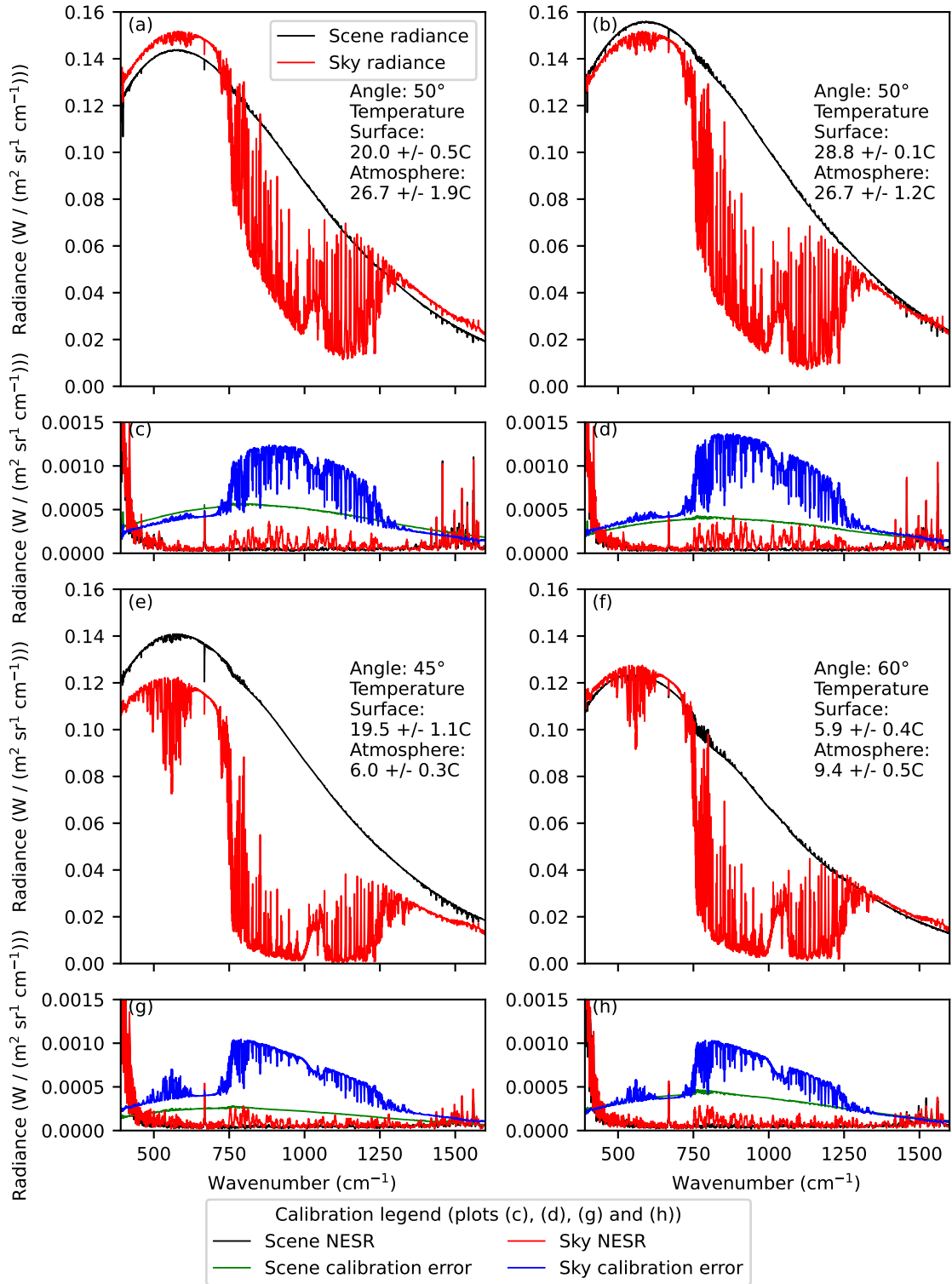


Figure 6.7: Spectra (a, b, e, f) and uncertainty (c, d, g, h) used in the emissivity retrievals for measurement sets (a,c) 1, (b,d) 2, (e,g) 3 and (f,h) 4. Each plot shows the average of 8 cycles corresponding to 1.5 hours of measurement time.

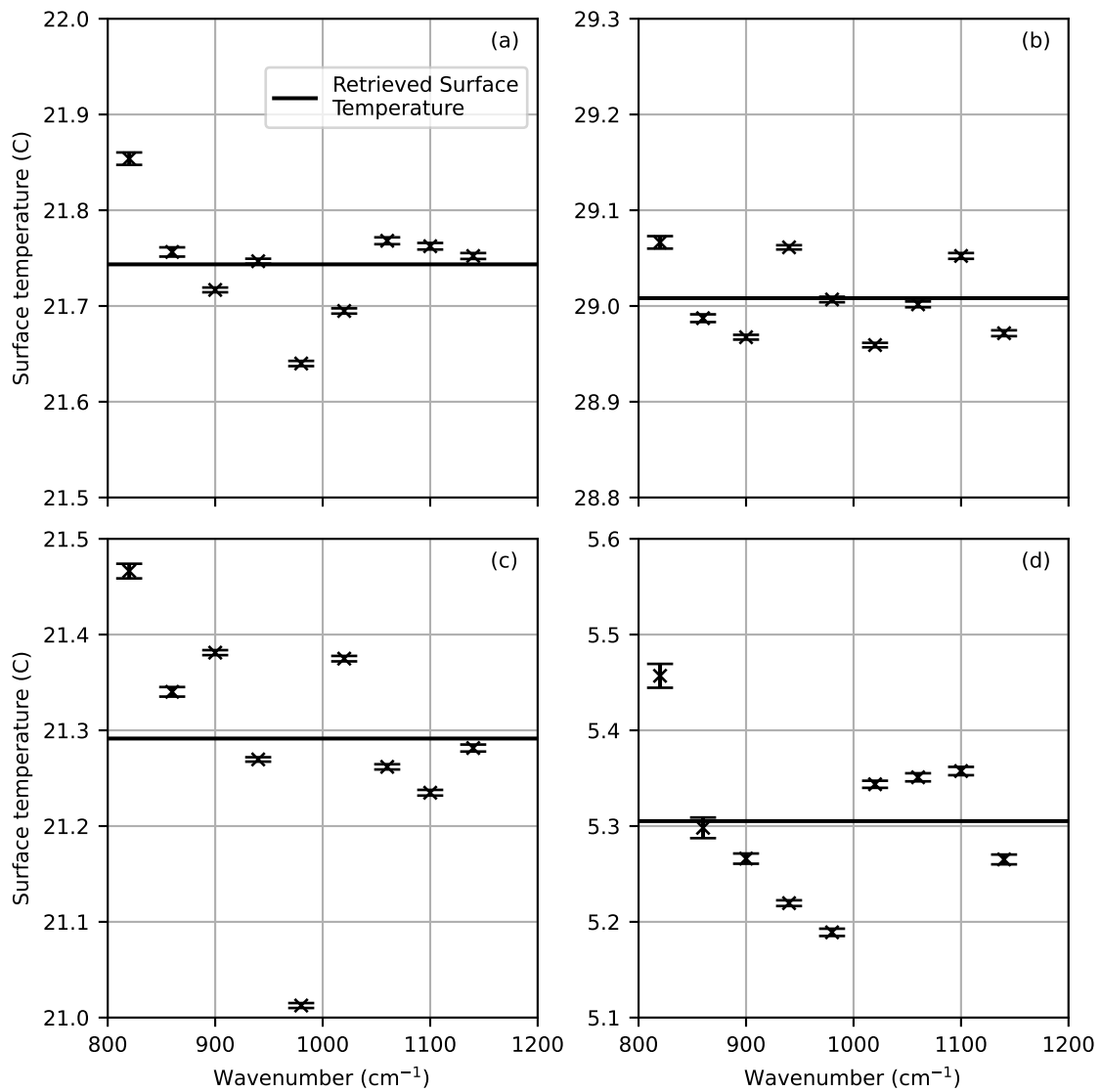


Figure 6.8: Examples of the retrieved surface temperature for each of the four sets of measurements. The black line is the average of the wavenumber bins and is taken to be the surface temperature.

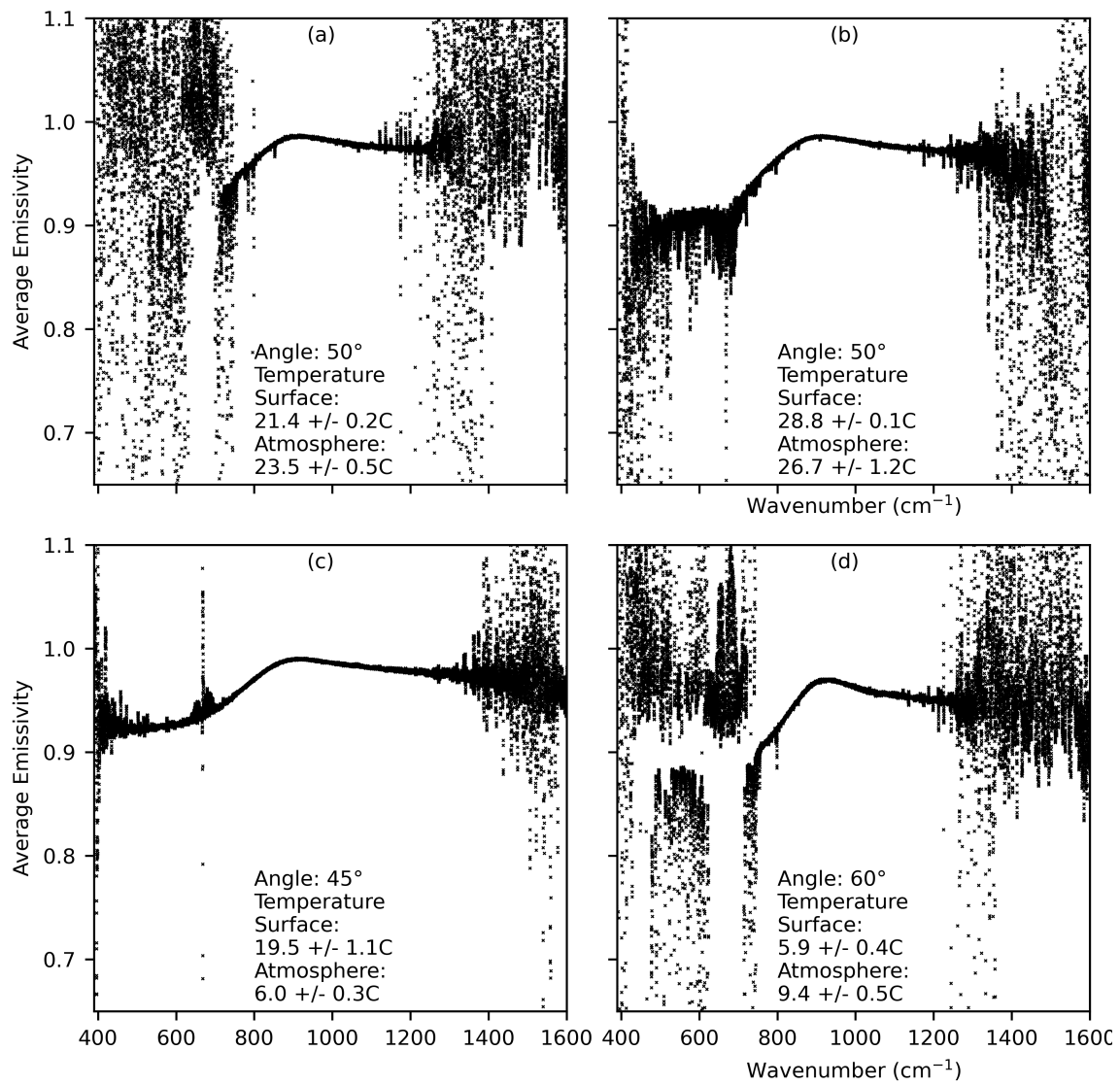


Figure 6.9: Initial emissivity retrievals for data from measurements (a) 1, (b) 2, (c) 3 and (d) 4. Each panel shows the average emissivity retrieved over 8 emissivity measurement cycles.

Figure 6.9 shows examples of the water emissivity retrieved using these spectra. Figure 6.9a shows data from the 1st set of measurements and figure 6.9b from the 2nd set of measurements. Both of these measurements were taken at an angle of 50° during the summer. Figure 6.9c shows data from the 3rd set of measurements which was taken at 45°. Figure 6.9d shows the data taken at 60° from the 4th set of measurements. Both the 3rd and 4th sets of data were taken in the winter. For the 2nd and 3rd sets of data the water was heated above the temperature of the atmosphere. Each plot shows the average of 8 cycles corresponding to 1.5 hours of measurement time. The effect of changing view angle can also be seen between 700 and 1000 cm^{-1} with larger angles having a steeper gradient. Figure 6.9a shows a large scatter in the data below 750 cm^{-1} and above 1300 cm^{-1} . Below 750 cm^{-1} there is some splitting of emissivity values with the lower, more plausible values seen just below 600 cm^{-1} in the regions of higher transmission. This is similar to the pattern seen with the FINESSE simulator in figure 5.16b and d indicating that this may be caused by a slight overestimation of the surface temperature. Figure 6.9b shows a lower scatter among the calculated points. As the emissivity is a smooth function of wavenumber, a lower scatter in the calculated emissivity is an indication of a better performing emissivity retrieval. The work with the FINESSE simulator shows that this improved performance is due to the increased surface temperature. The emissivity retrieval in figure 6.9c is the most successful, with the lowest scatter of points, particularly below 750 cm^{-1} . In this case this is due to the combination of the high surface temperature and low atmospheric temperature and water vapour concentration. The emissivity retrieval in figure 6.9d is also split between two values again indicating a slight overestimation of the surface temperature. Overall these measurements highlight the sensitivity of the retrieval to the derived surface temperature.

The data was then filtered using the contrast filter described in section 5.4.5 set at a contrast

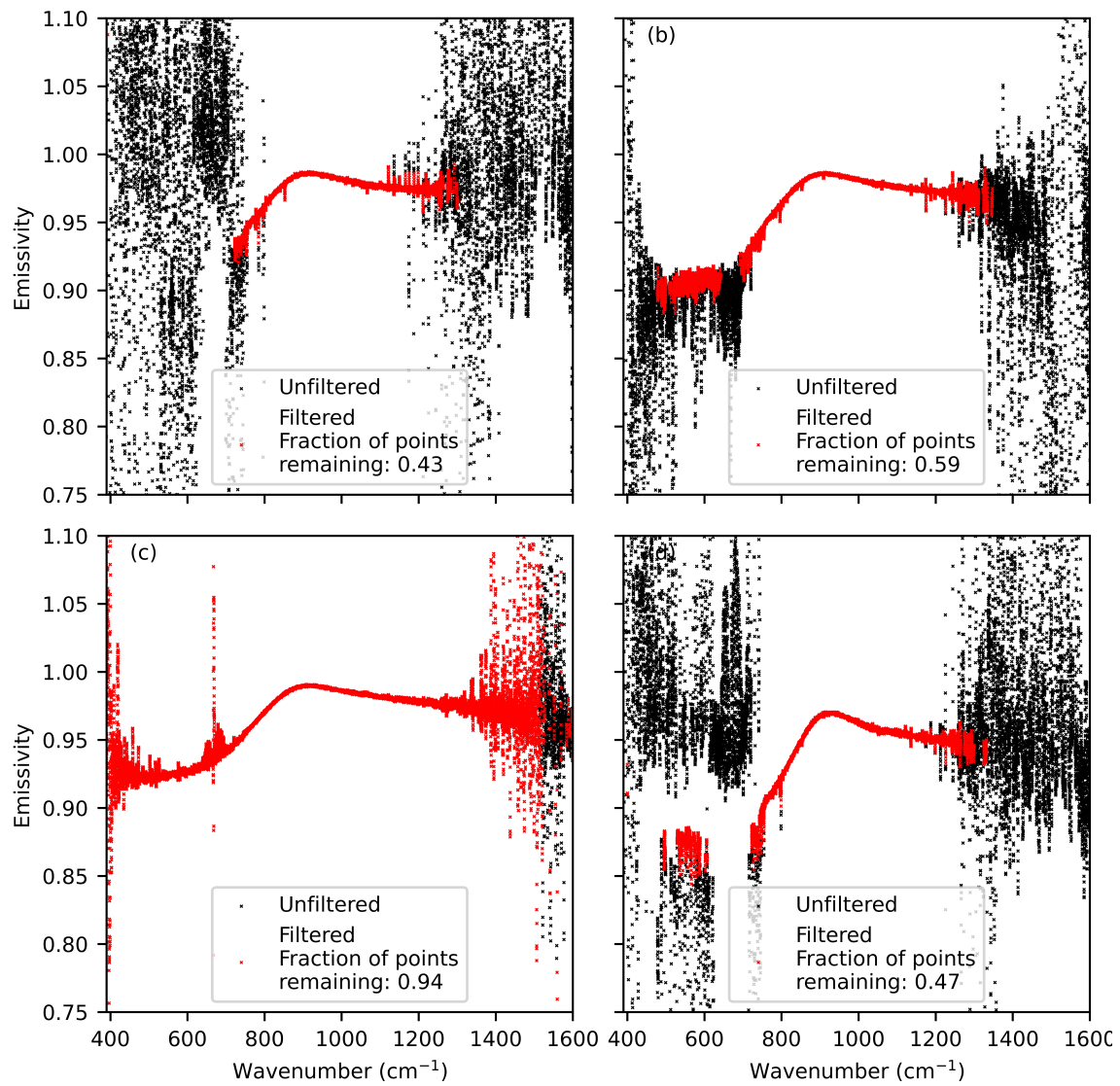


Figure 6.10: The same emissivity measurements as figure 6.9. The black points are unfiltered and the red points have been filtered using the contrast filter described in section 5.4.5.

level of $5 \text{ mW m}^{-2} \text{ sr}^{-1} \text{ cm}$. A comparison of the filtered and unfiltered data is shown in figure 6.10. In the case shown in figure 6.10a the contrast filter removes all the emissivity data below 700 cm^{-1} . This, coupled with the large scatter of points, indicates that the emissivity retrieval equation is very sensitive to uncertainty in input values and it is difficult to measure the emissivity in the FIR and above 1300 cm^{-1} when there is a warm, moist atmosphere present. This is expected given the work with the FINESSE simulator in section 5.4.5. The filter retains the largest fraction of points in the case shown in figure 6.10c when the surface is warmer than the atmosphere and the atmosphere is cool and contains less water vapour. The filter also identifies the more realistic emissivity values in figure 6.10d which is promising for the application of FINESSE to polar environments for the measurement of snow and ice emissivity. Despite retaining the most plausible values, the filter could still be improved. For example, some of the data from figure 6.10a could be useful. The filter also does not remove the data around the 667 cm^{-1} CO_2 band in figure 6.10c which is clearly affected by atmospheric features. After the data is filtered it is then spectrally averaged into bins of width 10 cm^{-1} . As the data was binned after filtering, the bins may contain different numbers of data points, particularly below 700 cm^{-1} . The standard deviation of the emissivity in each bin was also calculated. This binned emissivity is shown in figure 6.12 in comparison with theoretical calculations of the emissivity.

6.5 Comparison to Fresnel Calculations

The reflectance and hence emissivity of surfaces that act as specular reflectors can be calculated using Fresnel equations as long as the complex refractive indices are known (Masuda et al., 1988).

The reflectance, ρ , can be expressed as a function of viewing angle and refractive index

$$\rho(n, \theta) = \frac{(|\gamma_{\parallel}|^2 + |\gamma_{\perp}|^2)}{2}, \quad (6.1)$$

where n is the complex refractive index, θ is the viewing angle and $\gamma_{\parallel}$ and $\gamma_{\perp}$ are the complex reflectances polarized parallel and perpendicular to the plane of incidence, these are given by

$$\gamma_{\parallel} = -\frac{n \cos \theta - \cos \theta'}{n \cos \theta + \cos \theta'}, \quad (6.2)$$

$$\gamma_{\perp} = \frac{\cos \theta - n \cos \theta'}{\cos \theta + n \cos \theta'}, \quad (6.3)$$

where θ' is related to θ by

$$\sin \theta' = \frac{1}{n} \sin \theta. \quad (6.4)$$

The refractive index for pure water has been measured across the electromagnetic spectrum and compiled by various sources including Hale and Querry (1973), Downing and Williams (1975), Bertie and Lan (1996), and Max and Chapados (2009). Figure 6.11 shows the real and imaginary parts of these refractive indices. The differences between the different compilations are larger for the real part of the refractive index, particularly below 600 cm^{-1} . For the imaginary part, the largest differences are seen below 800 cm^{-1} . These compilations use data acquired using a range of measurement techniques over different wavenumber ranges and data from the same measurements are sometimes used in multiple compilations. The most recent compilation discussed in this thesis, Max and Chapados (2009), used new measurements in the wavenumber range $650 \text{ to } 6000 \text{ cm}^{-1}$ ($1.7 \text{ to } 15 \text{ }\mu\text{m}$). These data were taken using attenuated

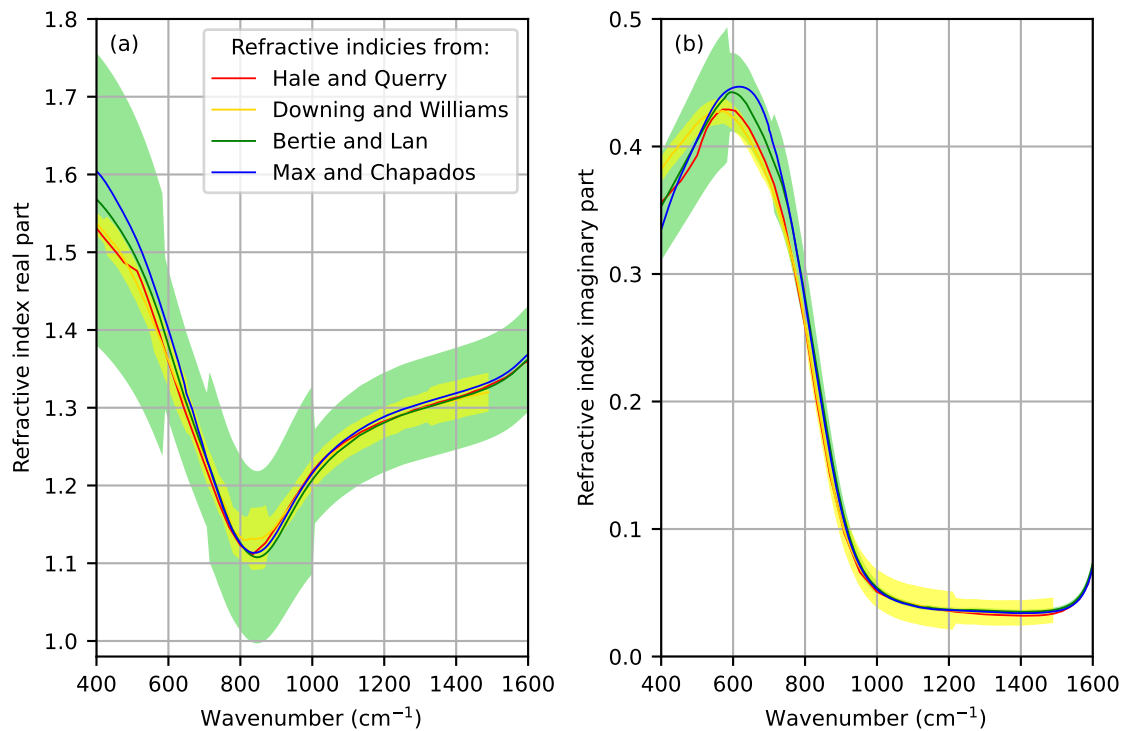


Figure 6.11: (a) real and (b) imaginary refractive indices from Hale and Query (1973), Downing and Williams (1975), Bertie and Lan (1996), and Max and Chapados (2009). The yellow and green shading represents the quoted uncertainty values from Downing and Williams (1975) and Bertie and Lan (1996).

total reflection measurements, which derive the refractive indices from measurements of how the intensity of radiation changes over multiple reflections within a sample (Bertie & Eysel, 1985). Below 650 cm⁻¹ (above 15 μm) the values from Bertie and Lan (1996) were combined with further new transmission measurements. These refractive indices are for a temperature of 25±2°C. Uncertainty values for these refractive indices are only given by Downing and Williams (1975) and Bertie and Lan (1996). These uncertainty values are shown as the yellow and green shading around the respective lines. For the real part of the refractive index and for wavenumbers below 800 cm⁻¹ in the imaginary part, the uncertainty estimate from the newer Bertie and Lan (1996) measurements is larger than that of the earlier Downing and Williams (1975) measurements. All values agree within the uncertainty values of the Bertie and Lan (1996) measurements. However it is not clear whether the earlier values of Hale and

Querry (1973) and Downing and Williams (1975) agree with the more recent values of Max and Chapados (2009) as these values do not have associated uncertainty estimates.

Figure 6.12 shows the filtered and spectrally averaged emissivity data from figure 6.10 with the theoretical emissivity calculated using Fresnel equations and the refractive indices from the four sources cited above. The uncertainty in the predicted emissivity caused by the quoted uncertainty in the refractive index is also shown. These calculations all assume a surface temperature of 25°C, despite this not being the case, as the refractive indices were measured at 25°C. The differences in the theoretical emissivity values seen between the panels is due to the different viewing angles for each case. Generally there is good agreement between the emissivity calculated using the Fresnel equations and the measured emissivity and the measured emissivity falls within the uncertainty bounds given by Bertie and Lan (1996). Above 900 cm^{-1} the theoretical emissivity values calculated using the four sets of refractive indices are very similar, and the differences between them are smaller than the standard deviation of the spectrally binned emissivity. Below 900 cm^{-1} the emissivity calculated using the newer refractive indices is lower than that calculated using the older refractive indices and this matches the measured emissivity more closely. Between 410 and 1300 cm^{-1} the standard deviation of the measured emissivity is lower than the uncertainty in the theoretical emissivity caused by the uncertainty in the Bertie and Lan (1996) refractive indices. This suggests that these measurements of water emissivity could be used to constrain the refractive index measurements as was done by Newman et al. (2005) in the MIR. The emissivity values between 400 and 650 cm^{-1} that are shown in figure 6.12b are lower than the values predicted using all sets of refractive indices, something that is not seen in panels c or d. Considering the sensitivity analysis in figure 5.18, this is consistent with an overestimation of the surface temperature of 0.02°C or the true downwelling spectrum being lower than the measured downwelling spectrum by an amount roughly 1.5 times

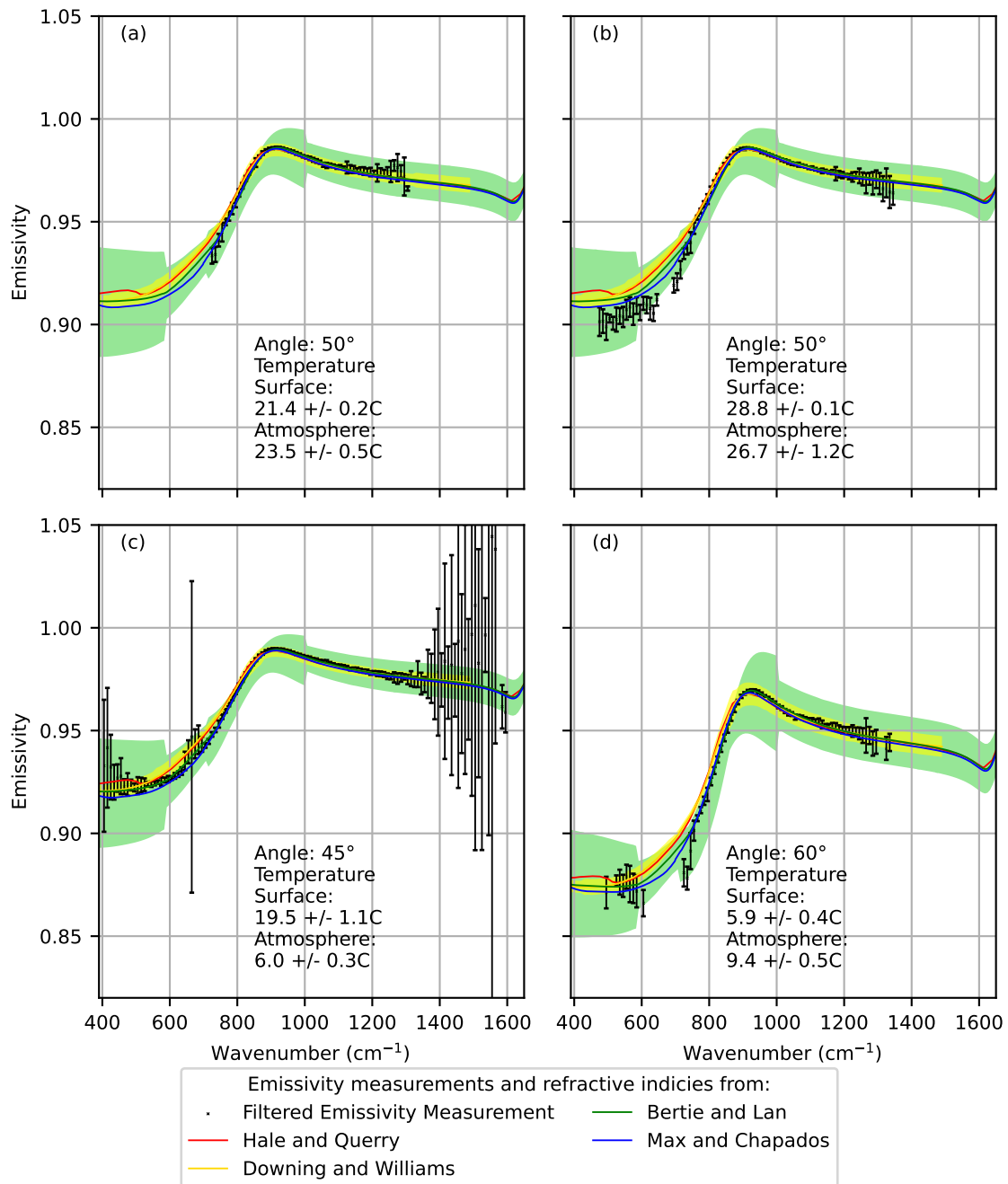


Figure 6.12: Filtered emissivity measurements and the emissivity calculated using Fresnel equations and the refractive indices from Hale and Query (1973), Downing and Williams (1975), Bertie and Lan (1996), and Max and Chapados (2009). The theoretical emissivity calculations assume a surface temperature of 25°C. The error bars show the standard deviation of the retrieved emissivity in each 10 cm⁻¹ bin.

the calibration error. This further highlights the sensitivity of the FIR emissivity retrieval to uncertainties in the input parameters, particularly surface temperature.

6.6 The Effect of Temperature on Emissivity Measurements

The refractive indices of water and hence the emissivity of water are dependent on temperature. From their measurements of water emissivity Newman et al. (2005) found that higher temperatures led to higher emissivity values between 750 and 1250 cm^{-1} . The location of the emissivity peak also shifted in wavenumber, moving to lower wavenumbers for higher temperatures.

Figure 6.13a shows the measured emissivity at an angle of 50° for surface temperatures of 28.8 and 5.0°C . Figure 6.13b shows the ratio between the two sets of emissivity measurements. The 28.8°C measurements are those shown above in figure 6.12b and the 5°C measurements were taken immediately before the measurements shown in figure 6.12d and are also averaged over 8 cycles. The measurements have been filtered and binned into 10 cm^{-1} bins. In agreement with the results of Newman et al. (2005), figure 6.13 shows a decrease in emissivity with decreasing temperature between 750 and 900 cm^{-1} . The change in emissivity is larger than the spread of emissivity values measured. Below 600 cm^{-1} , the emissivity values for the 5°C surface seem to increase in comparison with the 28.8°C values. However, the difference between the measurements is smaller than the scatter within the measurements so it is not possible to determine whether this is a real effect. Particular care should be taken with this comparison as figure 6.12b indicates that the 28.8°C measurements are lower than the theory suggests. A further experiment should be undertaken to measure emissivity as a function of temperature

systematically. This experiment could be carried out on a cold dry day with the water heated using the water bath in several temperature steps. To acquire more data the experiment could be repeated over several days with different atmospheric temperatures as when the water was heated too far above ambient temperature we observed the formation of mist which made the emissivity retrievals too difficult.

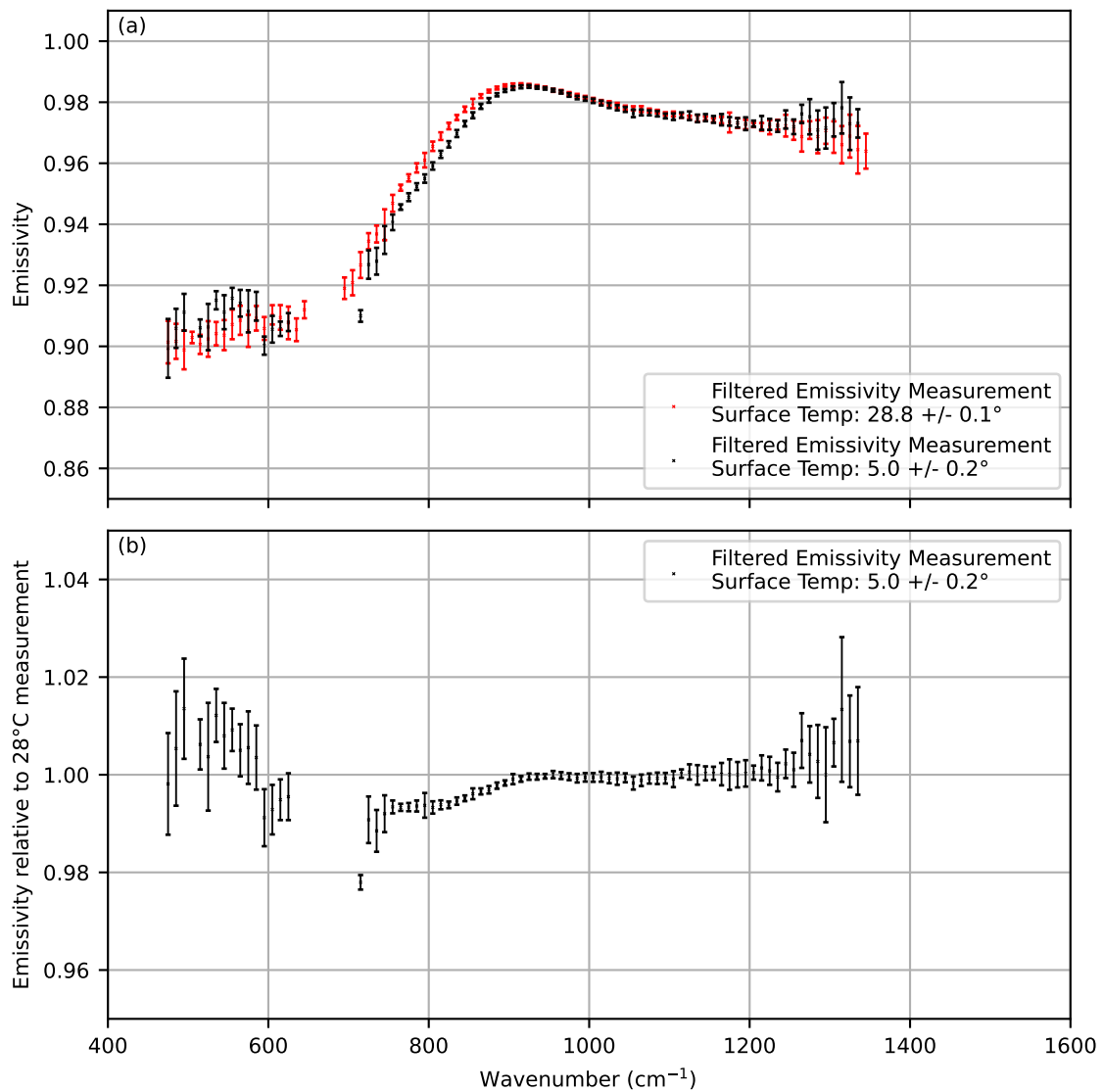


Figure 6.13: (a) Emissivity measurements for temperatures of 28.8°C (red) and 5.0°C (black). (b) Ratio between the emissivity measurements (5.0°C / 28.8 °C).

6.7 Angular Dependence of Emissivity

One of the key features of FINESSE is the steerable mirror which allows for straightforward measurements of emissivity at different angles. Therefore for the 4th set of emissivity measurements FINESSE undertook measurements of the emissivity of water at angles of 50°, 60° and 70°. Although not particularly representative of view angles from satellite measurements, these angles do cover the range at which the emissivity of water begins to vary dramatically with view angle as predicted by Fresnel calculations. This is therefore a suitable first test of the instrument and method.

Figure 6.14 shows the emissivity for the three angles compared to the emissivity calculated using Fresnel equations and the most recent set of refractive indices from Max and Chapados (2009). The measurements at each angle are the average of 8 cycles. These show generally good agreement with the predicted emissivity, the shape is consistent with predictions and the angular dependence is as expected. In the window region, the measurements at 50° agree best with the theoretical calculations, as the angle increases the measured emissivity becomes higher than the theoretical prediction. This is most noticeable around 900 cm⁻¹ in the measurements taken at 70°. It is more difficult to compare the theoretical calculations to the measurements below 900 cm⁻¹ because, as shown in section 6.6 and figure 6.13, the emissivity in this region is more strongly dependent on temperature. The average temperature of the water was 7°C during the measurements so figure 6.13 suggests that the retrieved emissivity values should be lower than the theoretical values due to the temperature mismatch. This is seen for the 50° retrieval but not the 60° or 70° retrievals. Below 600 cm⁻¹ the retrievals agree with the theoretical calculations although the spread of retrieved values is also larger. The longer path length between FINESSE and the surface makes the measurements at larger angles more difficult,

hence fewer points are retrieved for the 70° measurements. These measurements were also taken later in the day so the atmosphere was warmer possibly hindering the retrieval further. Despite these limitations, the angular measurements demonstrate the ability of FINESSE to make these important measurements.

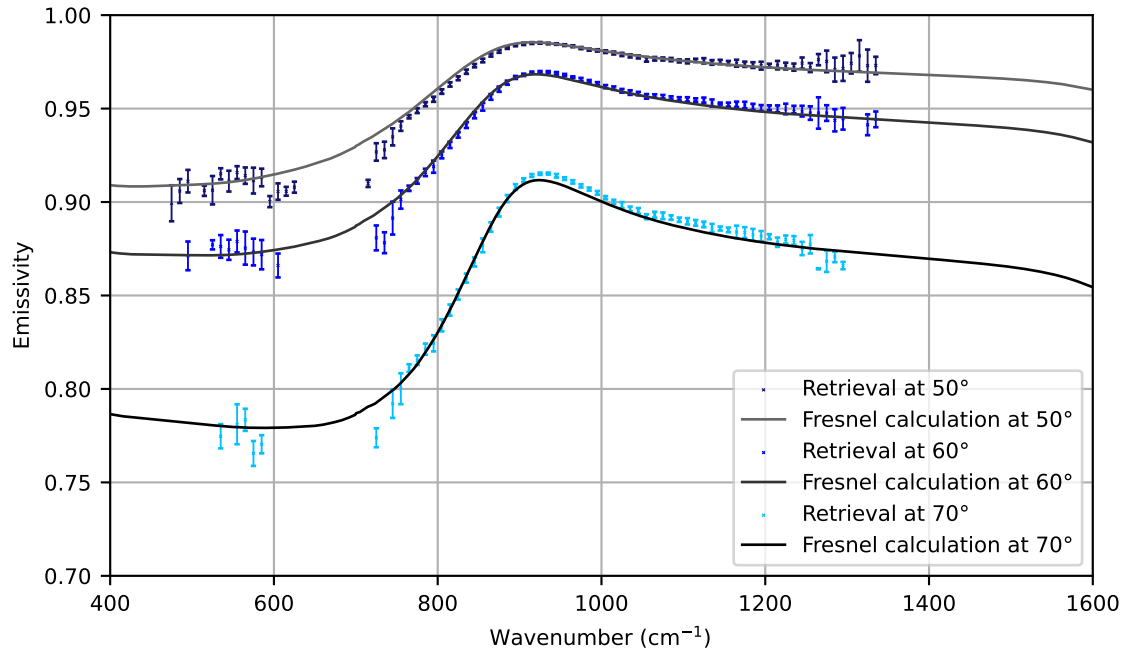


Figure 6.14: Emissivity measurements for three angles compared to the emissivity calculated using Fresnel equations and the refractive indices from Max and Chapados (2009).

6.8 Conclusion

This chapter has described some of the first emissivity measurements undertaken by FINESSE. These were of deionised water under a range of atmospheric conditions, surface temperatures and view angles. The method for calculating the emissivity was illustrated. As part of this the response function was calculated for each set of measurements and was shown to have good stability. However changing external conditions were shown to impact the response function over the course of several hours. These changes over a longer timescale were accounted for via the frequent calibration cycles undertaken. In the future a systematic study of the rate of

change of the response function could allow us to optimise the timing of the calibration cycles to take as much scene data as possible.

The retrieved emissivity was shown for a selection of experimental conditions. The emissivity retrieved using the test measurements showed features associated with the polypropylene view window. This motivated the change to a diamond view window which has a lower transmission but is without absorption features in the MIR and FIR. Of the emissivity retrievals carried out once the diamond window was installed, the most successful retrieval was the case with the cold dry atmosphere and the surface heated above the ambient air temperature. This is what was expected given the analysis completed with the FINESSE simulator. The filter proposed in section 5.4.5 was applied to the data and this successfully removed the non-physical results seen below 800 cm^{-1} however further work needs to be done to improve the data filtering further and to try and retain as much data as possible in the FIR. The emissivity retrieval was also spectrally averaged into bins of 10 cm^{-1} width. There are currently no published measurements of surface emissivity for water that extend below 667 cm^{-1} so these are novel measurements in their own right.

The retrieved emissivity was then compared to the emissivity calculated using Fresnel equations and refractive indices from a range of sources. All sets of refractive indices produced predicted emissivity values that matched the measurements in the MIR. In the FIR, differences in the predicted emissivity values became more apparent. However all sets of predicted emissivity values agreed within the uncertainty quoted by Bertie and Lan (1996) which was larger than the scatter in the retrieved emissivity. This suggests that emissivity measurements may usefully constrain the refractive index values. It should also be noted that the Huang emissivity database makes use of emissivity values from Hale and Querry (1973) which are very slightly higher than our measurements and do not have an associated uncertainty estimate. If this overestimation

is real then the true impact of FIR surface emissivity on the energy balance may be slightly larger than the current studies suggest.

The effect of temperature on emissivity values was then briefly investigated. In agreement with the MIR work of Newman et al. (2005), we found that the emissivity of water decreased with decreasing temperature between 700 and 900 cm^{-1} . The magnitude of this decrease was largest at 700 cm^{-1} where it had a value of 0.007. Below 600 cm^{-1} the scatter of retrieved emissivity values meant no definitive statements could be made about the effect of temperature on the emissivity. Future experiments to measure the effect of temperature on water emissivity below 600 cm^{-1} will be valuable in their own right but also important for future modelling and retrieval efforts. This is particularly relevant for retrievals from FORUM and PREFIRE as the ocean temperature of the water in polar regions where retrievals are likely to be possible will be below 25°C. To further aid the validation of the retrieval of the emissivity of ocean from satellite, measurements of the effect of salinity on emissivity should also be undertaken.

Finally, measurements were made of the emissivity of deionised water as a function of angle. These demonstrated the use of FINESSE's steerable mirror which made these measurements straightforward to undertake. In the MIR the retrieved emissivity values agreed with the theoretical predictions at 50°, however the measured emissivity was higher than predicted at 70°. In the FIR, the measured and theoretical emissivity values agreed within the scatter of the measurements. Going forwards, the method for making angular measurements of emissivity should be further refined, particularly for the measurement of diffuse rather than specular surfaces. The large angles measured and specular nature of the reflection from still water meant that the reflected downwelling radiance at the surface was represented by the downwelling radiance measured by FINESSE modulated only by the transmission and emission of the air between the surface and the instrument. However when measuring specular surfaces at small

angles, or when measuring diffuse surfaces such as snow, the reflected downwelling spectra may also be composed of radiation emitted by surrounding structures or FINESSE itself. Several techniques have been proposed to solve this problem, such as assuming that a certain proportion of the reflected downwelling radiance is composed of blackbody radiation at the temperature of the surrounding buildings (Borbas et al., 2021; Newman et al., 2005). A similar method will need to be developed for FINESSE.

These successful emissivity retrievals are a vital first step for FINESSE. Comparing measurements and theoretical predictions of a well characterised surface such as deionised water allows for the testing and refinement of the processes used to take measurements and analyse the results. This section has shown that it is possible to make FIR emissivity measurements using FINESSE and has highlighted several areas where further improvement is required. In order to further increase confidence in the measurements, a cross comparison could be made with other spectrometers. As FINESSE has a large range in the MIR between 667 and 1600 cm^{-1} (6.3 to $15\text{ }\mu\text{m}$), this cross comparison could be made with other MIR spectrometers such as ARIES.

The ultimate goal for the FINESSE instrument is deployment to make in-situ measurements of surface emissivity. Alongside the measurements already mentioned in this conclusion these measurements should include

- Measurements of sand. Sand is a diffuse reflector so measurements of sand emissivity at Imperial would be a convenient way to develop a method for making measurements of diffuse surfaces. Following the method of Bellisario et al. (2017) for snow surfaces, this method will likely involve assuming the sand is a Lambertian reflector and hence that the angularly integrated downwelling radiance can be approximated by measuring the downwelling radiation at an angle of around 55° . A method must also be developed to

take into account radiation emitted by FINESSE and other surrounding structures. These sand measurements are also of interest as Ben-Yami et al. (2022) have shown that it may be possible to make some retrievals of FIR emissivity of sandy surfaces using FORUM. These measurements will also enable validation of the Huang et al. (2016) data set.

- In-situ measurements of snow and ice emissivity. These measurements should be accompanied by snow characterisation including snow type and grain size. This is because measurements in the MIR (Hori et al., 2006) and FIR (Palchetti, Barucci, et al., 2020), and modelling in the far infrared (Huang et al., 2016) show that the emissivity is dependent on these properties, exhibiting differences of around 0.01 between 400-600 cm^{-1} . The analysis in section 5.4.5 suggests this is close to the size of the uncertainty in the emissivity retrieval. However the uncertainty in the retrieval can be reduced further by averaging. The uncertainty is also dependent on the measurement conditions and expected to be lower in colder, drier environments. Accurate measurements of snow and ice emissivity are particularly important because these surfaces tend to be present at the poles, where the atmosphere is less opaque to FIR radiation. This means the surface emissivity can possibly be retrieved from space (Ben-Yami et al., 2022; Xie et al., 2022) and also have a direct impact on the top of the atmosphere energy balance and a noticeable impact on the surface temperature produced by climate models (Kuo et al., 2018).
- Measurements of other natural surfaces, particularly those more likely to be found at high latitudes such as evergreen vegetation. Measurements of vegetation would be particularly valuable as these surfaces are very difficult to model and therefore the Huang emissivity database assumes the FIR emissivity of these surfaces is constant. However, it should be noted that the emissivity of vegetation is difficult to measure due to its complex structure so significant work would need to be undertaken to design a suitable measurement

strategy.

Overall, FINESSE has the potential to be used to make novel and impactful in-situ measurements of spectrally resolved emissivity in the FIR. The work completed in chapter 5 and this chapter are an important foundation on which these measurements can rest.

Chapter 7

Conclusions and Future Work

This thesis has described two separate strands of work exploring the use of spectrally resolved FIR radiance measurements to understand the vertical profile of atmospheric water vapour and to measure surface emissivity; the PIKNMIX-F campaign and the FINESSE instrument development.

Chapter 3 of this thesis described the PIKNMIX-F campaign which was undertaken in March 2019. During this campaign co-incident measurements of upwelling FIR and MIR radiance were undertaken. These measurements included clear sky cases and overflights of thin cirrus clouds. The data taken during this campaign are a valuable addition to the small number of airborne FIR measurements that existed before this campaign. The data have been used in two published studies: the work described in chapter 4, and Magurno et al. (2020) which describes an identification algorithm for cirrus clouds. Additional studies investigating water vapour spectroscopy and the radiative impacts of cirrus clouds are currently ongoing and there is further potential to utilise these data, for example in testing the end-to-end simulator that is being constructed for the FORUM mission.

In chapter 4, radiance measurements taken on a clear sky flight during the PIKNMIX-F campaign were used to investigate the sensitivity of the FIR spectrum to atmospheric water vapour. Forward modeling of the expected radiance at the aircraft level was carried out using LBLRTM and the weighting functions were calculated. The agreement between the simulations and measured radiances was generally good, however the agreement was less good in the TAFTS measurements between 425 and 550 cm^{-1} and in the ARIES measurements in the 667 cm^{-1} CO_2 band.

The discrepancy in the CO_2 band could be explained by local heating of the ARIES instrument housing. The difference in the TAFTS measurements between 425 and 550 cm^{-1} could not be completely explained by uncertainty in the surface conditions or the water vapour spectroscopy. This information was important for interpreting the water vapour and temperature retrievals which were carried out in collaboration with colleagues. The initial retrievals of the atmospheric water vapour profile, atmospheric temperature profile and skin surface temperature using FIR radiances did not perform well when compared to in-situ dropsonde measurements. The performance of the retrievals was greatly improved by removing the TAFTS measurements at wavenumbers above 440 cm^{-1} . These successful retrievals are the first published retrievals of temperature and water vapour profiles carried out using FIR radiances measured from aircraft. Compared to equivalent retrievals performed using MIR radiances measured by ARIES, the FIR retrievals had a higher number of DFS for both temperature and water vapour. The FIR retrievals also better matched the water vapour profile measured by the dropsonde. The advantage of the FIR for water vapour retrievals was maintained when the resolution of TAFTS was reduced to match that of ARIES and when the ARIES calibration error was reduced.

These results are observational confirmation of theoretical studies that demonstrate the enhanced performance of FIR radiances for the retrieval of atmospheric water vapour. This is

exciting for the possible performance of water vapour retrievals using measurements from FORUM. Due to the constraints of the field campaign the retrievals could only be performed in one set of atmospheric conditions. If data were available, it would be valuable to expand the analysis to a wider variety of atmospheric conditions. For example, in the theoretical analysis by Merrelli and Turner (2012) the FIR had the largest advantage compared to the MIR in the tropical profiles that were modelled.

The second half of the thesis focused on the development of FINESSE and its first measurements of the emissivity of water. Chapter 5 provided an introduction to the instrument including the calibration method and instrument line shape. The method for jointly retrieving surface temperature and emissivity was described. A FINESSE simulator was also developed. The simulator was used to assess the performance of the measurement of water emissivity under the conditions seen in London during the summer and winter. These simulations demonstrated that retrievals of surface emissivity in the FIR would be possible under these conditions. However the emissivity retrieval was much more sensitive to changes in measured radiance, atmospheric conditions and surface temperature at wavenumbers below 700 cm^{-1} , particularly when the contrast between the downwelling radiation and the surface emission was low. The surface temperature retrieval portion of the method performed well, however a key piece of future work should be the refinement of this method, particularly given the sensitivity of the retrieved emissivity values to the assumed surface temperature. The effect on the emissivity of perturbing the input parameters was also investigated using the simulator. In cases where the surface was warmer than the ambient air, the total effect of all these perturbations on the retrieved emissivity was around 1.5%. In cases where the surface was a similar temperature to the ambient air, the effect of the perturbations was much larger. However by filtering the data, regions could be isolated where the total effect of the perturbations was around 2%. The

simulator should have valuable applications as a tool for future work. For example it could be used in conjunction with climatological data to select locations and times for fieldwork which would result in the best emissivity measurements.

In chapter 6 FINESSE was used to make measurements of the emissivity of deionised water. Measurements were undertaken in summer and winter conditions, with the surface heated and unheated and at different angles. There are currently no published retrievals of the emissivity of water in the FIR so these measurements are a novel piece of work. During the course of analysing the measurements, the response function of FINESSE was investigated and found to have good stability, although it could be affected by the external conditions for example sharp changes in temperature or direct solar illumination. The FINESSE response function was also found to take an hour to stabilise, this was mainly due to the time taken for the HBB to reach a steady temperature. Therefore, since the measurements described in this thesis were made, more powerful heaters have been added to the HBB cavity.

The retrieved emissivity values showed good agreement with theoretical values calculated using Fresnel equations in both the MIR and FIR. This gives confidence to our method of retrieving surface emissivity and in the instrument. Future work should be done to develop data filtering or analysis methods to ensure the emissivity can be retrieved as successfully as possible in the FIR in cases where a moist and warm atmosphere is overlying a surface of a similar temperature. It would also be valuable to undertake cross-comparison with other spectrometers to further assess the performance of the instrument.

The effect of surface temperature on the emissivity of water was also briefly explored. In the MIR, the change in emissivity matched that seen in a previous study by Newman et al. (2005). However the range of data taken was not ideally suited for the detailed investigation

of temperature dependence and in the FIR the data were too noisy to draw any conclusions about the temperature dependence of the emissivity. More systematic measurements of the temperature dependence of emissivity should be carried out which will be valuable in their own right but also important for the validation of possible future emissivity retrievals from PREFIRE and FORUM.

Measurements of emissivity were also taken as a function of angle. These showed good agreement with theoretical values. The measurements were straightforward to undertake due to the steerable mirror included in the FINESSE calibration system. In the future this feature of FINESSE could be used to make measurements of the emissivity of a variety of surfaces as a function of angle.

Overall these first measurements made using FINESSE give us confidence in the instrument and method used to retrieve emissivity. Building on these measurements and the further work mentioned above, future work at Imperial should include the development of a method to measure diffuse surfaces such as sand. The ultimate aim for the FINESSE instrument is its deployment to make in-situ emissivity measurements. The theoretical work described in section 1.4.3 suggests that the first measurements should be of snow and ice emissivity accompanied by characterisation of the snow surface. However, due to the lack of measurements of FIR emissivity, measurements of a wide variety of surface types would be valuable. A more detailed list of future emissivity work can be found in section 6.8.

Outside of measurements of surface emissivity, measurements from FINESSE could also be used to investigate other aspects of FIR science. These could either be stand alone measurements or part of a larger field campaign. For example there is a possibility for FINESSE to take part in an ESA field campaign in support of the FORUM mission. The aim of the field

campaign would be to investigate the far-infrared signature of cirrus clouds and compare this to lidar measurements and simulations using radiative transfer models. This field campaign would involve FINESSE making measurements of downwelling radiance in the MIR and FIR alongside measurements from ground based lidar and co-incident in-situ sampling of the clouds by aircraft. These measurements would be valuable because Bantges et al. (2020) showed that current ice cloud models are not consistent across the whole infrared spectrum. Therefore more measurements of the radiative signature of ice clouds across the infrared in conjunction with in-situ cloud sampling are needed to address this difference. This particular field campaign is just one example of how FINESSE could be used outside measurements of surface emissivity. There are many possibilities for FINESSE to be used for exciting FIR science as it is well suited to making field measurements in a variety of locations due to being fairly portable and easy to operate; capable of variable viewing angles; and proven against theoretical predictions.

Appendix A

Derivatives of the Newman Emissivity Equation

This appendix contains the derivatives of the Newman emissivity retrieval equation (equation 5.13) with respect to the upwelling and downwelling radiance, transmission spectrum, atmospheric temperature and surface temperature.

The emissivity retrieval equation used in this thesis is given by

$$\epsilon = \frac{L_{det}^{\uparrow} - \tau^2 L_{det}^{\downarrow} - (1 - \tau^2)B(T_a)}{\tau\{B(T_s) - \tau L_{det}^{\downarrow} - (1 - \tau)B(T_a)\}} = \frac{g}{h} \quad (\text{A.1})$$

where ϵ is the emissivity, $L_{det}^{\uparrow}$ the upwelling radiation at the detector, $L_{det}^{\downarrow}$ the downwelling radiation at the detector, τ the transmission between the detector and the surface, $B(T_a)$ the Planck function at the temperature of the atmosphere and $B(T_s)$ the Planck function at the temperature of the surface Newman et al., 2005.

For ease of reference let

$$g = L_{det}^{\uparrow} - \tau^2 L_{det}^{\downarrow} - (1 - \tau^2)B(T_a) \quad (\text{A.2})$$

and

$$h = \tau\{B(T_s) - \tau L_{det}^{\downarrow} - (1 - \tau)B(T_a)\} \quad (\text{A.3})$$

Below are the derivatives of the Newman emissivity retrieval equation (equation 5.13) with respect to each parameter.

A.1 Upwelling Radiance

$$\frac{d\epsilon}{dL_{det}^{\uparrow}} = \frac{1}{h} = \frac{1}{\tau\{B(T_s) - \tau L_{det}^{\downarrow} - (1 - \tau)B(T_a)\}} \quad (\text{A.4})$$

A.2 Downwelling Radiance

$$\frac{d\epsilon}{dL_{det}^{\downarrow}} = \frac{L_{det}^{\uparrow} + (\tau - 1)B(T_a) - \tau B(T_s)}{[(\tau - 1)B(T_a) - \tau L_{det}^{\downarrow} + B(T_s)]^2} \quad (\text{A.5})$$

A.3 Transmission

$$\frac{d\epsilon}{d\tau} = \frac{g'(\tau)h(\tau) - g(\tau)h'(\tau)}{[h(\tau)]^2} \quad (\text{A.6})$$

where

$$g'(\tau) = 2\tau(B(T_a) - L_{det}^{\downarrow}) \quad \text{and} \quad h'(\tau) = 2\tau(B(T_a) - L_{det}^{\downarrow}) + B(T_s) + B(T_a) \quad (\text{A.7})$$

A.4 Atmospheric Temperature

$$\frac{d\epsilon}{dT_a} = \frac{d\epsilon}{dB(T_a)} \frac{dB(T_a)}{dT_a} \quad (\text{A.8})$$

where $\frac{dB(T)}{dT}$ is the derivative of the Planck function with respect to temperature. This expression is always positive and is given by

$$\frac{dB(T)}{dT} = \frac{2h^2c^3\nu^4 e^{\frac{h\nu}{k_bT}}}{k_bT^2(e^{\frac{h\nu}{k_bT}} - 1)^2} \quad (\text{A.9})$$

$$\frac{d\epsilon}{dB(T_a)} = \frac{g'(B(T_a))h(B(T_a)) - h'(B(T_a))g(B(T_a))}{[h(B(T_a))]^2} \quad (\text{A.10})$$

where

$$g'(B(T_a)) = \tau^2 - 1 \quad \text{and} \quad h'(B(T_a)) = \tau(\tau - 1) \quad (\text{A.11})$$

It should be noted that the transmission τ also changes when the atmospheric temperature changes but that is not included in this expression.

A.5 Surface Temperature

$$\frac{d\epsilon}{dT_s} = \frac{d\epsilon}{dB(T_s)} \frac{dB(T_s)}{dT_s} \quad (\text{A.12})$$

where

$$\frac{d\epsilon}{dB(T_s)} = \frac{-\epsilon}{B(T_s) - \tau L_{det}^\downarrow - (1 - \tau)B(T_a)} \quad (\text{A.13})$$

and $\frac{dB(T)}{dT}$ is defined in equation A.9.

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