Supplemental Information

Supplementary Figures

 Figure S1. Hourly DAC performance and ambient condition for Albuquerque, NW (cold dry climate) using 2020 data. Hourly production and electricity consumption assume steady production

equivalent to a large scale plant where process cycles for different collectors are synchronized.

 Figure S2. Hourly DAC performance and ambient condition for London, UK (cold humid climate) using 2020 data. Hourly production and electricity consumption assume steady production

equivalent to a large scale plant where process cycles for different collectors are synchronized.

Figure S3. Hourly DAC performance and ambient condition for Riyadh, SA (Hot dry climate) using

 2020 data. Hourly production and electricity consumption assume steady production equivalent to a 17 large scale plant where process cycles for different collectors are synchronized.

 Figure S4. Hourly DAC performance and ambient condition for Jakarta, ID (Hot humid climate) using 2020 data. Hourly production and electricity consumption assume steady production equivalent to a large scale plant where process cycles for different collectors are synchronized.

 Figure S5. Effect of relative humidity and temperature on productivity (tpaCO² per collector). It can 28 be seen that both temperature and relative humidity have an observable effect on process 29 productivity.

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 Figure S6. Effect of relative humidity and temperature on electricity requirement (MWhel per tCO2). It can be seen that relative humidity has a more pronounced effect (than temperature) on the total electricity requirement.

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 Figure S7. Thermal energy requirement breakdown. The thermal energy requirement for preheating the steam water supply is zero for all conditions as this heat requirement is met by the recovered heat from compressing the process gasses (Experimental Procedures).

 Figure S9. LCOD at different climates conditions and LCOEs. The variation in LCOD is due to the variation of regional DAC performance (due to the variation in regional climate) and WACC. SD is the standard deviation. Please find details about the calculation of standard deviation in Note S.7.

Figure S10**. Global DAC supply curves at different LOCEs when WACC is fixed at 5%.** Here, the

variation in LCOD is mainly due to the variation of DAC performance affected by regional climate.

⁶³ Supplementary Tables

64 **Table S1. Economic analysis assumptions.**

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66 $^{\circ}$ ^a All costs are adjusted to 2020 using CE index = 596.2.

67 bThe CO₂ collector cost can be estimated based on the similarity of the materials used to manufacture 68 it compared to cars^{3,4}, including steel, aluminium, plastics, insulation, etc. Similar to a car, the CO₂ 69 collector is modular; therefore, it can be manufactured using similar process methods used for car 70 manufacturing. Moreover, a material assessment of the $CO₂$ collector can estimate the amount of 71 materials needed for each collector. Each collector has 6 square-shaped steel walls, two of which are 72 movable, with each side having dimensions of around 1.45 m. Assuming a wall thickness of 0.01 m, 73 the total steel weight needed is 990 kg at a density of 7850 kg per $m³$. Each collector has 88 frames. 74 Each frame consists of 21.3 m of aluminium tubes with a diameter of 0.01 m and a thickness of 0.001 75 m. Thus, each collector contains 242 kg of aluminium tubes at a density of 2710 kg per $m³$. Aluminium 76 mesh is used to improve the bed thermal conductivity where each frame contains 1.9 kg of aluminium 77 mesh based on the ratio used in Climeworks patent³, resulting in an additional 166.5 kg of aluminium 78 per collector. Additional aluminium and plastic are used for C-profiles, sturts, wedge spacers and 79 sealing strips at a ratio of 0.53 kg per frame or 46.5 kg per collector. Based on this calculation, the 80 material needed for one collector is around 1446 kg per collector, including steel, aluminium and 81 plastic. This can be compared to a mid-sized car's average weight of around 1500 kg (ref⁵).

82 CAPEX (Experimental Procedures).

83 Heat pumps are estimated based on the main process equipment (compressors) cost, then

84 adjusted to account for the total installed cost, including auxiliary equipment using Lang Factor

85 (Experimental Procedures).

⁸⁶ Note S1: adsorption model parameters

- 87 The table below summarizes the model parameters used for the adsorption model.
- 88 Adsorption model parameters.

89 The concentration of H₂O varies based on the relative humidity.

90 b The concentration of air varies based on the concentration of H₂O which calculated as

91 $1 - y_{CO2} - y_{H2O}$

93 The following are some comments regarding the model parameters.

94 • The reported sorbent envelope density is 630–710 kg m⁻³ (Young el al.⁷). 635.2 kg m⁻³ is used 95 here as the envelope density to simplify the analysis. At this envelope density, each $CO₂$ 96 collector contains 1000 kg of sorbent. Solid density (ρ_s) , envelope density (ρ_e) , and bulk 97 density (ρ_b) are defined in Equations 1–3 where V_T , V_x , V_p , and M are total system volume, 98 external void volume, internal void volume (pore volume), and mass.

$$
\rho_s = \frac{M_{particles}}{V_T - V_X - V_P} \tag{1}
$$

$$
\rho_e = \frac{M_{particles}}{V_T - V_X} \tag{2}
$$

$$
\rho_b = \frac{M_{particles}^{'}}{V_T} \tag{3}
$$

- 99
- 100 When calculating the bed porosity, it was assumed that the aluminium mesh occupies around 101 3.8% of the sorbent cell, based on an estimate from the patent³.
- 102 The reported particle diameter for the modelled sorbent is in the range of 3.15×10^{-4} 103 1.25×10⁻³ m (Young el al.⁷). 7.5×10⁻⁴m is used, which is around the average, as at this particle 104 diameter, the pressure drop for 1×10^{-2} m thickness bed is around 1 mbar when air velocity is 105 0.028 m s⁻¹ This pressure drop at this air velocity is aligned with the data presented in the 106 patent³.
- 107 The thermal conductivity of the sorbent bed was estimated from the patent testing 3 . In the 108 testing, a sorbent cell similar to the one modelled here was heated by flowing a heating fluid 109 through the heating tube with a temperature of 93.6 °C, with the centre of the bed reaching 110 a temperature of 85 °C after 90 minutes. Process condition information such as the sorbent 111 condition and desorption pressure for the testing was not reported in the patent. It was 112 assumed here that the sorbent was saturated with $CO₂$ and H₂O under an ambient condition 113 of 20°C and RH of 40%. Two desorption pressures were used to estimate the thermal 114 conductivity: 1000 mbar and 100 mbar. The figure below shows the model fit on the patent 115 testing data and the estimated thermal conductivity for the two desorption pressures which 116 is in the range of 0.42–0.72 W m⁻¹ K⁻¹. It was not possible to obtain an exact fit, mainly due 117 to the missing process information from the patent. However, in both fits, the bed centre 118 temperature reached around 85 °C within 90 minutes. The sorbent used in the patent was 119 amine-functionalized cellulose which has a thermal conductivity of 44.5×10^{-3} W m⁻¹ k^{-1 8}. 120 Considering the calculated thermal conductivity (0.42-0.72 W m⁻¹ K⁻¹), this corresponds to an 121 improvement between 9−16 times the thermal conductivity of cellulose by incorporating the 122 aluminium mesh design, which is probably reasonable given that aluminium thermal 123 conductivity is 205 W m⁻¹ K⁻¹. For Lewatit VP OC 1065, the thermal conductivity is 0.16 W m⁻¹ 124 K^{-1} (Young el al.⁷). It is assumed that the thermal conductivity of the modelled bed using 125 Lewatit VP OC 1065 is the average of the fitted range, which is 0.57 W m⁻¹ K⁻¹. This implies 126 that the thermal conductivity of the bed did not improve when Lewatit VP OC 1065, with 127 better thermal conductivity, was used instead of the cellulose sorbent. Also, thermal 128 conductivity ofthe bed has been improved by 3.6 times compared to the Lewatit VP OC 1065 129 thermal conductivity when incorporating the aluminium mesh. This might be in the 130 conservative range given that the thermal conductivity of aluminium is 3 orders of magnitude 131 larger than Lewatit VP OC 1065.

Thermal conductivity patents³ 133 **data fitting.**

137 • In Wurzbacher et al.⁸ work, it was found that diffusion has a strong effect on the temperature 138 profile of the process especially when operating at vacuum, as diffusion is inversely 139 proportional to pressure. However, their simplified approach using an average value for the 140 3-component mixture of CO₂, water and air did not affect the accuracy of the modelling. 141 Therefore, the same approach is adopted in this work.

¹⁴² Note S2: Isotherm model parameters

143 The following two tables show the isotherm model parameters that are used in this work which is 144 based on Young el al.⁷

Mechanistic and WADST co-adsorption model fitting parameters⁷ 145 **.**

GAB model fitting parameters⁷ 147 **.**

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¹⁴⁹ Note S3: Adsorption model boundary conditions

150 The boundary conditions for all simulated cyclic steps are shown in table below where N is the number 151 of finite volume cells. \dot{F} , A, y_i , P and p are volumetric flow rate, cross sectional area in the flow 152 direction, gas concentration of component I, total pressure and partial pressure, respectively. x , y and 153 $-z$ are variables for space as shown in Figure 7. In the 1D model, $\frac{1}{2}$ represents the boundary wall of 154 bhe first finite volume cell, which is equivalent to $z = 0$, and $N + \frac{1}{2}$ represents the boundary wall 155 of the last finite volume cell, which is equivalent to $z = l$.

 For each cyclic step, the initial conditions for the variables are the final values of the variables from the previous step. When there is a transition from the 1D model to the 2D model, the variable average over the z-axis or x-y plane in case of the 1D model or 2D model, respectively, from the previous step 159 is used as an initial condition for the current step. k_{BL} corresponds to the valve equation controlling 160 the pressure where a value of 0.25 s⁻¹ is used. More detailed of each cyclic step and process description is presented in Note S4.

162 **Adsorption model boundary conditions.**

¹⁶⁴ Note S4: Steam-assisted vacuum-pressure temperature swing ¹⁶⁵ adsorption process description

167 **A systematic of SA-VTSA cycle.**

168 The figure above shows the cyclic design for the SA-TVSA cycle, where all the boxes represent the 169 same CO₂ collector. In the adsorption step, fans push the air through the collector, where CO₂ is 170 adsorbed into the sorbent along with some H₂O. After the sorbent has reached the desired $CO₂$ 171 saturation (97%), the collector is sealed and evacuated to vacuum (50 mbar) using a vacuum pump 172 (i.e., blowdown step). This step is relatively fast, and the sorbent does not desorb a significant amount 173 of CO₂ at this point, and thus mostly air is removed, which is vented back to the atmosphere. Removing 174 the air from the contactor before heating the sorbent is important in case of utilizing amine-175 functionalised solid sorbents. This is because the amines in the sorbent can have critical degradation 176 when exposed to oxygen at high temperature (i.e., oxidative degradation)⁹⁻¹¹. For the desorption step, 177 the sorbent is heated using a heating fluid at 100 °C using the heat transfer tubes, leading to the 178 desorption of CO₂ and water, which are removed instantaneously due to the vacuum. After all points 179 on the sorbent bed reach at least 95 °C, the heating fluid is stopped and the $CO₂$ collector is purged 180 with steam (at steam gas velocity ($u_{inlet\ stream}$) of 0.01 m s⁻¹). The steam purging lowers the CO₂ partial 181 pressure inside the collector, leading to an increase of the $CO₂$ working capacity. The steam purging 182 step stops when the bed reach an average $CO₂$ loading of 0.1 mol $CO₂$ per kg sorbent. After the sorbent 183 is regenerated, the bed is cooled down using a cooling fluid with a temperature above ambient 184 temperature by 10 °C using the heat transfer tubes. The cooling step stops when all points on the 185 sorbent are colder than 65 °C. Then, the collector is opened back to the atmosphere, and the cycle is 186 repeated. The cooling step is important as the amine groups on the sorbent might get oxidized when 187 the hot sorbent is contacted with oxygen in the air $9-11$.

S.17

188 Supplementary Note $SS: CO₂$ compression modelling

190 **Process flow diagram of the CO² compression section.**

 The process section shown in the figure above is modelled using a combination of Aspen Plus V11 and MATLAB R2021a. Aspen Plus is used to provide fluid properties calculation where Lee Kesler Pokler (LKP) Equation of State (EoS) was selected as it was found to be suitable for the CO₂ and H₂O mixture¹². MATLAB is used to automate the process synthesis as the section feed and operating parameters change due to changes in ambient conditions and feed conditions. In the automated process synthesis, 196 HX-1 only recovers heat if the feed inlet temperature for HX-1 ($T_{inlet,HX-1}$) is above the heating fluid 197 temperature $T_{heating}$ by the minimum approach temperature ΔT_{min} , which is 10°C in this work. HX-2 198 recovers the remaining heat and cools down the stream to ΔT_{min} above the ambient temperature T_{amb} such that CO₂ does not change phase to the liquid phase. When the feed stream contains mostly water, HX-2 recovers a substantial amount of low-temperature heat that is caused by condensing water, which cannot be recovered for preheating the steam water supply. Therefore, when accounting for the amount of heat that can be recovered from HX-2, if the inlet stream has a water mole fraction of more than 0.5, the heat is neglected and not recovered. The remaining amount of heat from HX-2 is recovered at 80% efficiency to account for any remaining water phase change.

205 The table below shows the algorithm used to set the parameters of the two heat exchangers where 206 P_{sat,CO_2} and T_{sat,CO_2} are saturation vapor pressure and its temperature (i.e., boiling temperature). A 207 bigher temperature than CO₂ critical temperature and T_{sat,CO_2} are used as design margin.

208 **An algorithm for the selection of operating parameter in the CO² compression section.**

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²¹⁰ Note S6: Model validation

211 Climeworks reports the productivity of their $1st$ generation collector of 50 ktaCO₂ per collector¹³ (ambient conditions were not reported for this collector productivity). Reported energy 213 requirements¹⁴ is based on daily averaged data from the two Climeworks plants (Hellisheidi, Iceland and Hinwil, Switzerland). It is not clear whether Climeworks uses steam purging in these plants (like 215 the model in our paper), however, Climeworks do hold a patent for process configurations using steam 216 purging after heating of the bed¹⁵. A comparison of the reported Climeworks data with the model 217 results is summarised below in the table below. It is worth noting that energy requirements here are reported as separate thermal energy and electricity requirements. The thermal energy requirements include heat required for heating the bed (by the heating fluid) and steam generation, whereas electricity requirements include electricity needed for vacuum pumps compressors and fans. In the main paper, it is assumed that heat pumps that consume electricity are used to provide the thermal energy requirements needed to heat the bed (by the heating fluid) and generate steam. Thus, the reported electricity requirements in the main paper includes the heat pumps.

224 **Comparison of the model results with Climeworks data.**

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226 There is reasonable agreement between the model estimates of process productivity and thermal 227 energy requirement with reported data by Climeworks for real-world units operating in two locations

 (with temperatures below zero). However, for electricity requirements, there is a quite considerable 229 deviation which was noticed before by Sabatino et al.¹⁶. The potential reasons for deviation may include sorbent type used and its weight per collector, process configuration, if steam purging or heat integration are used, and what is included in electricity requirements (e.g., building electricity, lights 232 etc.) as well as vacuum pumps, compressors and fans efficiencies. Also, the electricity requirements 233 for fans are compared to the data provided in the Climeworks patent³. Climeworks reported 0.08 234 MWh_{el} per tCO₂ at a fan efficiency of 70%, a pressure drop of 1 mbar through the adsorption bed, and a capture rate of 70%. Although we assume the same fan efficiency and target the same pressure drop, the calculated capture rate from the model is higher, which explains the slightly lower electricity 237 requirements for fans of 0.06 MWh_{el} per $tCO₂$.

 Thermal energy requirements include heat required for heating the bed (by the heating fluid) and steam calculated from the model.

 Electricity requirements include electricity required for fans, vacuum pumps, and compressors calculated from the model.

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²⁵⁶ Note S7: standard deviation calculation

257 The standard deviation is calculated using MATLAB (2021a) function std. Since not all spatial nodes

258 has the same area, weighted standard deviation is used where node DAC performance is weighted

259 by the node area. The MATLAB std function uses the following relation:

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S_{w} = \sqrt{\frac{\sum_{i=1}^{N} w_{i} |A_{i} - \mu_{w}|^{2}}{\sum_{i=1}^{N} w_{i}}}
$$

261

$$
\mu_{w} = \frac{\sum_{i=1}^{N} w_i A_i}{\sum_{i=1}^{N} w_i}
$$

262 where

- A_i each value from the population,
 N size of the population.
- size of the population,
- μ_{w} weighted mean,
- S_w weighted standard deviation,
- w weighting scheme.

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