# Atmospheric energy budget response to idealized aerosol perturbation in tropical cloud systems

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# 11 Abstract

The atmospheric energy budget is analysed in numerical simulations of tropical cloud systems 12 13 to better understand the physical processes behind aerosol effects on the atmospheric energy 14 budget. The simulations include both shallow convective clouds and deep convective tropical 15 clouds over the Atlantic Ocean. Two different sets of simulations, at different dates (10-12/8/2016 and 16-18/8/2016), are simulated with different dominant cloud modes (shallow or 16 17 deep). For each case, the cloud droplet number concentrations (CDNC) is varied as a proxy for changes in aerosol concentrations without considering the temporal evolution of the aerosol 18 19 concentration (for example due to wet scavenging which may be more important under deep convective conditions). It is shown that the total column atmospheric radiative cooling is 20 21 substantially reduced with CDNC in the deep-cloud dominated case (by  $\sim 10.0 \text{ W/m}^2$ ), while a much smaller reduction (~1.6 W/m<sup>2</sup>) is shown in the shallow-cloud dominated case. This trend 22 23 is caused by an increase in the ice and water vapor content at the upper troposphere that leads to a reduced outgoing longwave radiation, an effect which is stronger under deep-cloud dominated 24 conditions. A decrease in sensible heat flux (driven by increase in the near surface air 25 temperature) reduces the warming by  $\sim 1.4 \text{ W/m}^2$  in both cases. It is also shown that the cloud 26 fraction response behaves in opposite ways to an increase in CDNC, showing an increase in the 27 deep-cloud dominated case and a decrease in the shallow-cloud dominated case. This 28 demonstrates that under different environmental conditions the response to aerosol perturbation 29 could be different. 30

32 Introduction

The negative anthropogenic radiative forcing due to aerosols is acting to cool the climate and to compensate some of the warming due to increase in greenhouse gases (Boucher et al., 2013). However, quantification of this effect is highly uncertain with a revised uncertainty range of -1.60 to -0.65 W/m<sup>2</sup> (Bellouin et al., 2019). The total anthropogenic aerosol radiative forcing is composed of contribution from direct interaction of aerosols with radiation (scattering and absorption) and from indirect interaction with radiation due to changes in cloud properties.

39 Beside its effect on the radiation budget, aerosols may affect the precipitation distribution and total amount (Levin and Cotton, 2009; Albrecht, 1989; Tao et al., 2012). A useful perspective to 40 41 improve our understanding of aerosol effect on precipitation, which became common in the last 42 few years, arises from constraints on the energy budget (O'Gorman et al., 2012; Muller and O'Gorman, 2011; Hodnebrog et al., 2016; Samset et al., 2016; Myhre et al., 2017; Liu et al., 43 44 2018; Richardson et al., 2018; Dagan et al., 2019a). On long time scales, any precipitation perturbations by aerosol effects will have to be balanced by changes in radiation fluxes, sensible 45 heat flux or by divergence of dry static energy. The energy budget constraint perspective was 46 found useful to explain both global (e.g. (Richardson et al., 2018)) and regional (Liu et al., 2018; 47 48 Dagan et al., 2019a) precipitation response to aerosol perturbations in global scale simulations. In this study, we investigate the energy budget response to aerosol perturbation on a regional 49 50 scale using high resolution cloud resolving simulations. This enables an improved understanding 51 of the microphysical processes controlling atmospheric energy budget perturbations. The strong 52 connection between the atmospheric energy budget and convection has long been appreciated (e.g. (Arakawa and Schubert, 1974; Manabe and Strickler, 1964)) as well as the connection to 53 54 the general circulation of the atmosphere (Emanuel et al., 1994).

55 The total column atmospheric energy budget can be described as follows:

56  $LP + Q_R + Q_{SH} = \operatorname{div}(s) + \operatorname{ds/dt}$  (1)

Equation 1 presents a balance between the latent heating rate (*LP* - latent heat of condensation [*L*] times the surface precipitation rate [*P*]), the surface sensible heat flux ( $Q_{SH}$ ), the atmospheric radiative heating ( $Q_R$ ), the divergence of dry static energy (div(*s*), which will become negligible on sufficiently large spatial scales), and the dry static energy storage term (ds/dt, which will

become negligible on long [inter-annual] temporal scales). Throughout the rest of this paper we will refer to the right-hand side of Equation 1 (div(s)+ds/dt) as the energy imbalance (which is calculated as the residual [R] of the left-hand side).

64  $Q_R$  is defined as:

65  $Q_R = (F_{SW}^{TOA} - F_{SW}^{SFC}) + (F_{LW}^{TOA} - F_{LW}^{SFC})$  (2)

and represents the rate of net atmospheric diabatic warming due to radiative shortwave (SW) and
longwave (LW) fluxes. It is expressed by the sum of the surface (SFC) and top of the atmosphere
(TOA) fluxes, when all fluxes are positive downwards. As in the case of TOA radiative forcing,
aerosols could modify the atmospheric energy budget by both direct interaction with radiation
and by microphysical effects on clouds. The latter is the focus of this study.

The microphysical effects are driven by the fact that aerosols serve as cloud condensation nuclei 71 72 (CCN) and ice nuclei (IN). Larger aerosol concentrations, e.g. by anthropogenic emissions, could 73 lead to larger cloud droplet and ice particle concentrations (Andreae et al., 2004; Twomey, 1977; Hoose and Möhler, 2012). Changes in hydrometer concentration and size distribution were 74 75 shown to affect clouds' microphysical processes rates (such as condensation, evaporation, freezing and collision-coalescence), which in turn could affect the dynamics of the clouds (Khain 76 77 et al., 2005; Koren et al., 2005; Heikenfeld et al., 2019; Chen et al., 2017; Altaratz et al., 2014; Seifert and Beheng, 2006a), the rain production (Levin and Cotton, 2009; Albrecht, 1989; Tao 78 et al., 2012) and the clouds' radiative effect (Koren et al., 2010; Storelvmo et al., 2011; Twomey, 79 1977; Albrecht, 1989). The aerosol effect, and in particular its effects on the radiation budget 80 and the atmospheric energy budget, is cloud regime dependent (Altaratz et al., 2014; Lee et al., 81 2009; Mülmenstädt and Feingold, 2018; van den Heever et al., 2011; Rosenfeld et al., 2013; 82 Glassmeier and Lohmann, 2016; Gryspeerdt and Stier, 2012; Christensen et al., 2016), time 83 84 dependent (Dagan et al., 2017; Gryspeerdt et al., 2015; Seifert et al., 2015; Lee et al., 2012; Dagan et al., 2018c), aerosol type and size distribution dependent (Jiang et al., 2018; Lohmann 85 and Hoose, 2009) and (even for a given cloud regime) meteorological conditions dependent 86 (Dagan et al., 2015a; Fan et al., 2009; Fan et al., 2007; Kalina et al., 2014; Khain et al., 2008) 87 and was shown to be non-monotonic (Dagan et al., 2015b; Jeon et al., 2018; Gryspeerdt et al., 88 2019; Liu et al., 2019). Hence the quantification of the global mean radiative effect is extremely 89 challenging (e.g. (Stevens and Feingold, 2009; Bellouin et al., 2019)). 90

91 Previous studies demonstrated that the mean aerosol effect on deep convective clouds can increase the upward motion of water, and hence also increase the cloud anvil mass and extent 92 (Fan et al., 2010; Chen et al., 2017; Fan et al., 2013; Grabowski and Morrison, 2016). The 93 increase in mass flux to upper levels was explained by the convective invigoration hypothesis 94 95 (Fan et al., 2013; Koren et al., 2005; Rosenfeld et al., 2008; Seifert and Beheng, 2006a; Yuan et al., 2011a; Williams et al., 2002), which was proposed to lead to stronger latent heat release 96 under higher aerosol concentrations and hence stronger vertical velocities. In addition to the 97 stronger vertical velocities, under polluted conditions the smaller hydrometers are being 98 99 transported higher in the atmosphere (for a given vertical velocity (Chen et al., 2017; Koren et al., 2015; Dagan et al., 2018a)) and their lifetime at the upper troposphere is longer (Fan et al., 100 101 2013; Grabowski and Morrison, 2016). The invigoration mechanism can also lead to an increase 102 in precipitation (Khain, 2009; Altaratz et al., 2014). Both the increase in precipitation and the increase in anvil coverage would act to warm the atmospheric column: the increased precipitation 103 by latent heat release, and the increased anvil mass and extent by longwave radiative warming 104 (Koren et al., 2010; Storelvmo et al., 2011). However, it should be pointed out that the 105 uncertainty underlying these proposed effects remain significant (White et al., 2017; Varble, 106 107 2018). In addition, aerosol effects on precipitation from deep convective cloud was shown to be 108 non-monotonic and depend on the aerosol range (Liu et al., 2019).

In the case of shallow clouds, aerosol effect on precipitation was also shown to be non-monotonic 109 (Dagan et al., 2015a; Dagan et al., 2017). However, unlike in the deep clouds case, the mean 110 effect on precipitation, under typical modern-day conditions, is thought to be negative (Albrecht, 111 1989; Rosenfeld, 2000; Jiang et al., 2006; Xue and Feingold, 2006; Dagan and Chemke, 2016). 112 The aerosol effect on shallow cloud cover and mean water mass (measure by liquid water path -113 LWP) might also depend on the meteorological conditions and aerosol range (Dagan et al., 114 2015b; Dagan et al., 2017; Gryspeerdt et al., 2019; Dey et al., 2011; Savane et al., 2015) and is 115 116 the outcome of competition between different opposing response of: rain suppression (that could lead to increase in cloud lifetime and coverage (Albrecht, 1989)), warm clouds invigoration (that 117 118 could also lead to increase in cloud coverage and LWP (Koren et al., 2014; Kaufman et al., 2005; Yuan et al., 2011b)) and increase in entrainment and evaporation (that could lead to decrease in 119 cloud coverage (Small et al., 2009; Jiang et al., 2006; Costantino and Bréon, 2013; Seigel, 120 2014)). Another addition to this complex response is the fact that the aerosol effect on warm 121 convective clouds was shown to be time dependent and affected by the clouds' feedbacks on the 122 thermodynamic conditions (Seifert et al., 2015; Dagan et al., 2016; Dagan et al., 2017; Lee et al., 123

2012; Stevens and Feingold, 2009; Dagan et al., 2018b). Previous simulations that contained 124 several tropical cloud modes demonstrate that increase in aerosol concentrations can lead to 125 suppression of the shallow mode and invigoration of the deep mode (van den Heever et al., 2011). 126 Hence the domain mean effect, even if it is demonstrated to be small, may be the result of 127 opposing relatively large contributions from the different cloud modes (van den Heever et al., 128 129 2011). The small domain mean effect may suggest that on large enough scales the energy (Muller and O'Gorman, 2011; Myhre et al., 2017) or water budget (Dagan et al., 2019b) constrain 130 precipitation changes. 131

Previous studies, using global simulations (O'Gorman et al., 2012; Muller and O'Gorman, 2011; Hodnebrog et al., 2016; Samset et al., 2016; Myhre et al., 2017; Liu et al., 2018; Richardson et al., 2018; Dagan et al., 2019a), demonstrated the usefulness of the atmospheric energy budget perspective in constraining aerosol effect on precipitation. However, the physical processes behind aerosol-cloud microphysical effects on the energy budget are still far from being fully understood. In this study we use cloud resolving simulations to increase our understanding of the effect of microphysical aerosol-cloud interactions on the atmospheric energy budget.

## 139 Methodology

140 The icosahedral nonhydrostatic (ICON) atmospheric model (Zängl et al., 2015) is used in a limited area configuration. ICON's non-hydrostatic dynamical core was evaluated with several 141 142 idealized cases (Zängl et al., 2015). The simulations are conducted such that they are aligned with the NARVAL 2 (Next-generation Aircraft Remote-Sensing for Validation Studies (Klepp 143 et al., 2014; Stevens et al., 2019; Stevens et al., 2016)) campaign, which took place during August 144 2016 in the western part of the northern tropical Atlantic. We use existing NARVAL 2 145 convection-permitting simulations (Klocke et al., 2017) as initial and boundary conditions for 146 our simulations. 147

The domain covers ~22° in the zonal direction (25° - 47° W) and ~11° in the meridional direction (6° - 17° N) and therefore a large fraction of the northern tropical Atlantic (Fig. 1). During August 2016, the intertropical convergence zone (ITCZ) was located in the southern part of the domain while the northern part mostly contains trade cumulus clouds. Hence, this case study provides an opportunity to study heterogenous clouds systems. Daily variations in the deep/shallow cloud modes in our domain were observed, but it always included both cloud modes, albeit in different relative fraction. Two different dates are chosen, one representing a shallow-cloud dominated 155 mode (10-12/8/2016 - see Fig. 2, and Figs S1 and S3, supporting information- SI), and one that represents a deep-cloud dominated mode (16-18/8/16 - see Fig. 3 and Figs. S2 and S3, SI). In 156 the shallow-cloud dominated case, most of the domain is covered by trade cumulus clouds that 157 are being advected with the trade winds from north-east to south-west. In the southern part of the 158 domain, throughout most of the simulation, there is a zonal band of deep convective clouds (Fig. 159 2) that contribute on average ~25% out of the total cloud cover (Fig. S3, SI). The deep-cloud 160 dominated case represents the early stages of the development of the tropical storm Fiona (Fig. 161 3). Fiona formed in the eastern tropical Atlantic and moved toward the west-north-west. It started 162 163 as a tropical depression at 16/8/2016 18:00 UTC while its centre was located at 12.0° N 32.2° W. It kept moving towards the north-west and reach a level of a tropical storm at 17/8/2016 12UTC, 164 located  $13.7^{\circ}$ Ν while its centre was at 36.0° W 165 (https://www.nhc.noaa.gov/data/tcr/AL062016\_Fiona.pdf). The general propagation speed and 166 direction, strength (measure by maximal surface wind speed) and location of the storm are 167 predicted well by the model. However, the model produces more anvil clouds than what was 168 observed from the satellite (Fig. 3). These two different cases, representing different atmospheric 169 energy budget initial state (see also Figs. 4 and 12 below), enable the investigation of the aerosol 170 effect on the energy budget under different initial conditions. 171

We use a two-moment bulk microphysical scheme (Seifert and Beheng, 2006b). For each case, 172 four different simulations with different prescribed cloud droplet number concentrations 173 (CDNC) of 20, 100, 200, and 500 cm<sup>-3</sup> are conducted. The different CDNC scenarios serve as 174 a proxy for different aerosol conditions (as the first order effect of increased aerosol 175 concentration on clouds is to increase the CDNC, Andreae, 2009). This also allows to separate 176 the cloud response from the uncertainties involved in the representation of the aerosols in 177 numerical models (Ghan et al., 2011; Simpson et al., 2014; Rothenberg et al., 2018). However, 178 it limits potential feedbacks between clouds and aerosols, such as the removal of aerosol levels 179 by precipitation scavenging and potential aerosol effects thereon. In addition, the fixed CDNC 180 framework does not capture the differences in aerosol activation between shallow and deep 181 clouds, due to differences in vertical velocity. Another aerosol effect that is not included in our 182 simulations is the direct interaction between aerosol and radiation. In future work we plan to 183 184 examine the mutual interaction between the microphysical effects and the direct aerosol radiative effects. 185

186 For calculation of the difference between high CDNC (polluted) conditions and low CDNC (clean) conditions, the simulations with CDNC of 200 and 20 cm<sup>-3</sup> are chosen as they represent 187 the range typically observed over the ocean (see for example the CDNC range presented in 188 recent observational-based studies (Rosenfeld et al., 2019; Gryspeerdt et al., 2019)). Each 189 190 simulation is conducted for 48 hours starting from 12 UTC. The horizontal resolution is set to 1200 m and 75 vertical levels are used. The temporal resolution is 12 sec and the output interval 191 192 is 30 min. Interactive radiation is calculated every 12 min using the RRTM-G scheme (Clough et al., 2005; Iacono et al., 2008; Mlawer et al., 1997). We have added a coupling between the 193 194 microphysics and the radiation to include the Twomey effect (Twomey, 1977). This was done by including the information of the cloud liquid droplet effective radius, calculated in the 195 microphysical scheme, in the radiation calculations. No Twomey effect due to changes in the 196 197 ice particles size distribution was considered due to the large uncertainty involved in the ice microphysics and morphology. Additional details, such as the surface and atmospheric physics 198 parameterizations, are described in Klocke et al., (2017) and include an interactive surface flux 199 200 scheme and fixed sea surface temperature (SST). We note that using a fixed SST does not 201 include feedbacks of aerosols on the SST evolution that could change the surface fluxes. However, due to the large heat capacity of the ocean, we do not expect the SST to dramatically 202 203 change over the two days simulations.

For comparing the outgoing longwave flux from the simulations and observations we use 204 205 imager data from the SEVIRI instrument onboard the Meteosat Second Generation (MSG) 206 geostationary satellite (Aminou, 2002). The outgoing longwave flux is calculated using the 207 Optimal Retrieval for Aerosol and Cloud (ORAC) algorithm (Sus et al. 2017; McGarragh, et al. 2017). Cloud optical (thickness, effective radius, water path) and thermal (cloud top 208 209 temperature and pressure) properties are retrieved from ORAC using an optimal estimationbased approach. These retrievals and reanalysis profiles of temperature, humidity and ozone 210 are then ingested into BUGSrad, a two-stream correlated-k broadband flux algorithm (Stephens 211 et al., 2001) that outputs the fluxes at the top and bottom of the atmosphere and shown to have 212 excellent agreement when applied to both active (CloudSat) and passive (Advanced Along 213 Track Scanning Radiometer) satellite sensors compared to Clouds and the Earth's Radiant 214 215 Energy System (Henderson et al. 2013; Stengel et al. 2019). In addition, off-line sensitivity radiative transfer tests using vertical profiles from our model were conducted with BUGSrad 216 to identify the source of the differences in fluxes between clean and polluted conditions. 217

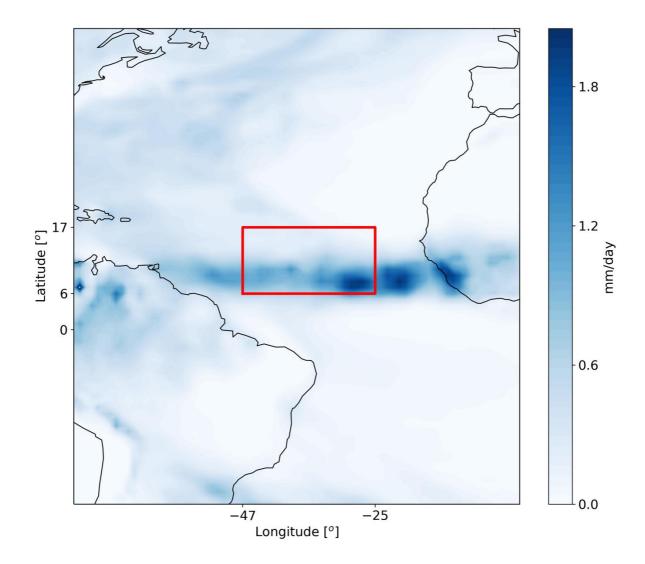


Figure 1. Domain of the ICON simulations (red rectangle) overlaid on the August 2016 ECMWF erainterim reanalysis (Dee et al., 2011) mean precipitation rate.

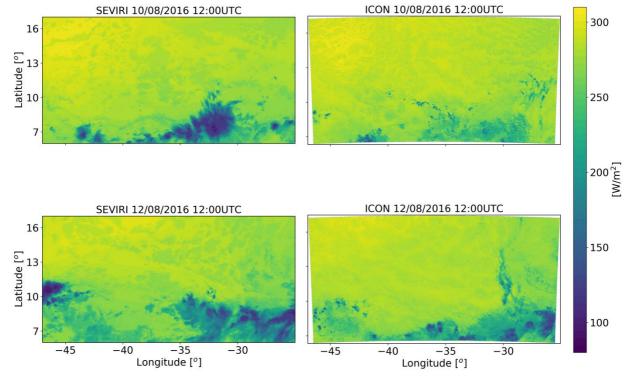
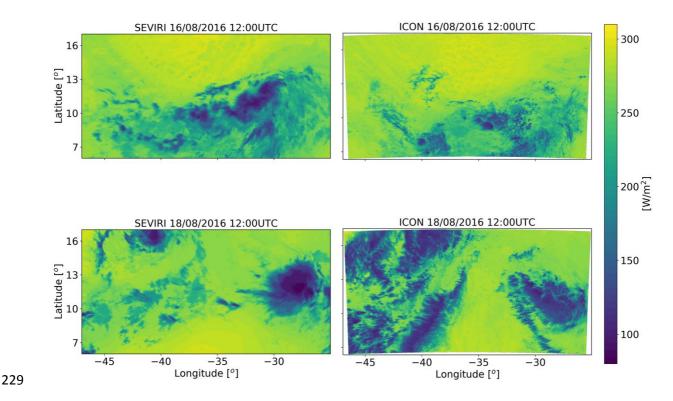


Figure 2. Outgoing longwave flux at the top of atmosphere at the initial stage (upper row) and the last stage (lower row – each average over 30 minutes) of the simulation of the shallow-cloud dominated case (10-12/08/2016) from geo-stationary satellite (SEVIRI-MSG – right column) and the ICON model simulation with CDNC of 20 cm<sup>-3</sup> (left column).

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230 Figure 3. similar to Figure 2 but for the deep-cloud dominated case (16-18/08/2016).

231 **Results** 

### 232 Shallow-cloud dominated case -10-12/08/2016

We start with energy budget analysis of the shallow-cloud dominated case base simulations 233  $(CDNC = 20 \text{ cm}^{-3})$ . Figure 4 presents the time mean (over the two days simulation) of the 234 different terms of the energy budget (Equation 1). As expected, LP dominates the warming of 235 236 the atmosphere while  $Q_R$  dominate the cooling. The sensible heat flux ( $Q_{SH}$ ) is positive (act to warm the atmosphere) but it is an order of magnitude smaller than the LP and  $Q_R$  magnitudes. In 237 this shallow-cloud dominated case the radiative cooling of the atmosphere is significantly larger 238 than the warming due to precipitation (mean of -114.7  $W/m^2$  compared to 90.1  $W/m^2$ ), hence the 239 energy imbalance (R) is negative. Negative R means that there must be some convergence of dry 240 static energy into the domain and/or decrease in the storage term, in this case it is mostly due to 241 convergence of dry static energy. 242

243 We note that there is a significant difference in the spatial distribution of LP and  $Q_R$  (Jakob et

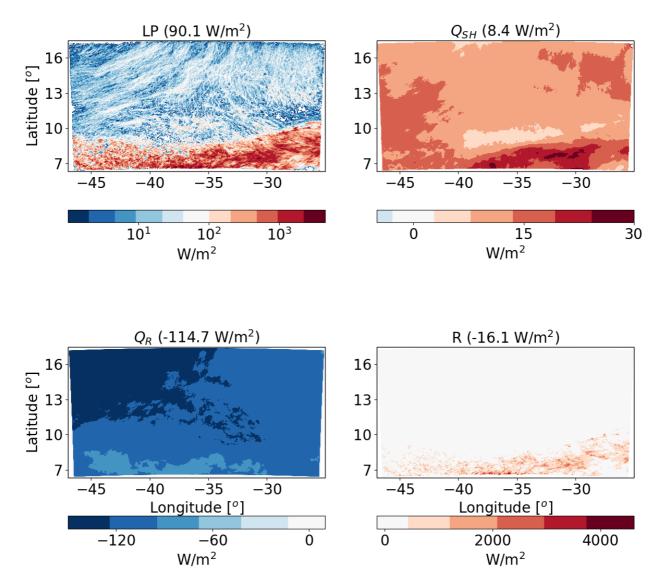
al., 2019). While the  $Q_R$  is more uniformly distributed, the LP is mostly concentrated at the south

245 part of the domain (where the deep convective clouds are formed) and it has a dotted structure.

Locally, at the core of a deep convective clouds, the *LP* contribution can reach a few  $1000 \text{ W/m}^2$ 

247 (1 mm/hr of precipitation is equivalent to  $628 \text{ W/m}^2$ ), however, the vast majority of the domain

contributes very little in terms of *LP*. *Q<sub>R</sub>* also presents some spatial structure in which there is a
weak atmospheric cooling at the south part of the domain (the region of the deep convective
clouds) and a strong cooling at the reset of the domain.



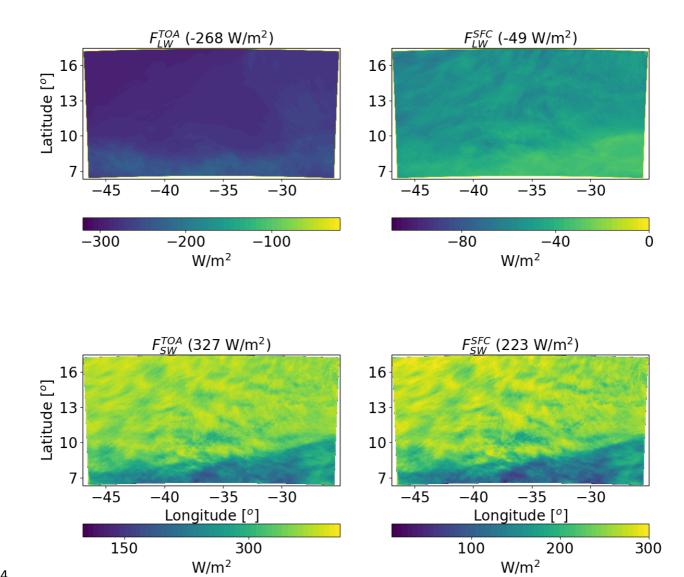
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Figure 4. Spatial distribution of the time mean of the different terms of the energy budget for the ICON simulation of the shallow-cloud dominated case (10-12/08/2016) with CDNC = 20 cm<sup>-3</sup>. The terms that appear here are: *LP* - latent heat by precipitation,  $Q_{SH}$  - sensible heat flux,  $Q_R$  - atmospheric radiative warming, and R – the energy imbalance. The domain and time-mean value of each term appears in parenthesis.

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For understanding the spatial structure of  $Q_R$ , next we examine the spatial distribution of the LW and SW radiative fluxes at the TOA and surface (Fig. 5). We note that the smaller radiative cooling in the region of deep clouds in the south of the domain is mostly contributed by a decrease in  $F_{LW}^{TOA}$ . The SW fluxes also demonstrate a strong south-north gradient, as the deep

convective clouds in the south are more reflective than the shallow trade cumulus (with the lowermean cloud fraction) in the rest of the domain.



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Figure 5. Spatial distribution of ICON simulated time-mean longwave (LW) and shortwave (SW) radiation fluxes at the top of atmosphere (TOA) and surface (SFC) for a simulation of the shallow-cloud dominated case (10-12/08/2016) with CDNC = 20 cm<sup>-3</sup>. The domain and time mean value of each term appears in parenthesis.

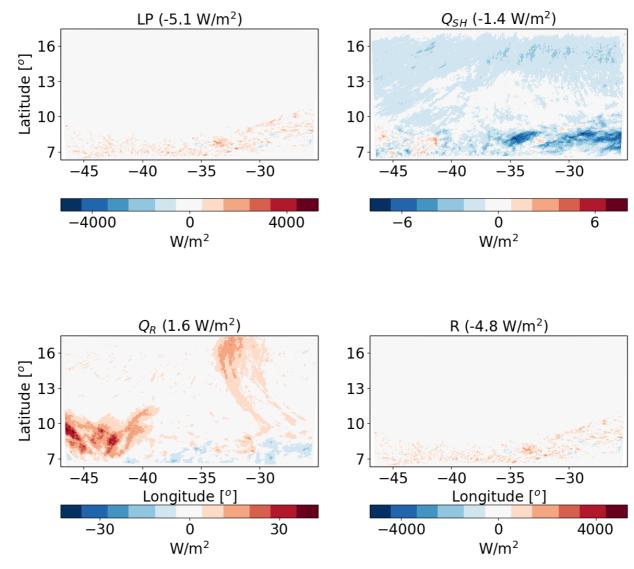
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## 270 Response to aerosol perturbation – shallow-cloud dominated case

Next, we analyse the response of the atmospheric energy budget of this case to perturbations in

272 CDNC. Figure 6 presents the differences in the different terms of the energy budget between a

polluted simulation (CDNC =  $200 \text{ cm}^{-3}$ ) and a clean simulation (CDNC =  $20 \text{ cm}^{-3}$ ). It 273 demonstrates that the LP differences between the different CDNC scenarios contribute 5.1 W/m<sup>2</sup> 274 less to warm the atmosphere in the polluted vs. the clean simulation. We note that this apparently 275 276 large effect is caused by a small, non-statistically significant, precipitation difference (~0.4 mm 277 over the two days of simulation - see Fig. 8 below). The strong sensitivity of the atmospheric energy budget to small precipitation changes (recalling that 1 mm/hr is equivalent to  $628 \text{ W/m}^2$ ) 278 279 exemplifies the caution one needs to take when looking on precipitation response in terms of energy budget perspective. The  $Q_R$  differences lead to relative warming of the atmosphere of the 280 polluted case compared to the clean case by 1.6  $W/m^2$ . We note that most of the  $Q_R$  differences 281 are located in the south-west part of the domain. The  $Q_{SH}$  changes counteracts 1.4 W/m<sup>2</sup> of the 282 atmospheric warming by  $Q_R$  and so the end result is a deficit of 4.8 W/m<sup>2</sup> in the atmospheric 283 energy budget in the polluted simulation compared to the clean simulation. The decrease in the 284 *Q<sub>SH</sub>* is driven by an increase in the near surface air temperature (see Fig. 8). 285



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Figure 6. The differences between polluted (CDNC = 200 cm<sup>-3</sup>) and clean (CDNC = 20 cm<sup>-3</sup>) ICON simulations of the time-mean terms of the energy budget for the shallow-cloud dominated case (10-12/08/2016). The terms that appears here are: *LP* - latent heat by precipitation,  $Q_{SH}$  - sensible heat flux,  $Q_R$  - atmospheric radiative warming, and R – the energy imbalance. The domain and time mean value of each term appears in parenthesis.

To understand the response of  $Q_R$  to the CDNC perturbation, we next examine the response of 293 the different radiative fluxes. Figure 7 demonstrates that most of the relative atmospheric 294 radiative heating in the polluted case compared to the clean case is contributed by changes in the 295  $F_{LW}^{TOA}$  fluxes. The changes in  $F_{LW}^{SFC}$  are an order of magnitude smaller. The SW fluxes change both 296 at the TOA and SFC are larger than the  $F_{LW}^{TOA}$  changes, however, in terms of the atmospheric energy 297 budget, they almost cancel each other out and the net SW atmospheric effect is only  $-0.9 \text{ W/m}^2$ . 298 299 Most of the reduction in SW fluxes (both at TOA and the surface) comes from the deep 300 convective regions in the south of the domain while the shallow cloud regions experience some

increase in SW fluxes. This can be attributed to the increase in deep convective cloud fraction
 and a decrease in the shallow cloud fraction with the increase in CDNC (see Fig. 9 below). The
 TOA net radiative effect for the entire system (as opposed to the atmospheric energy budget that
 take into consideration the surface radiative fluxes changes) is about -5.2 W/m<sup>2</sup>.

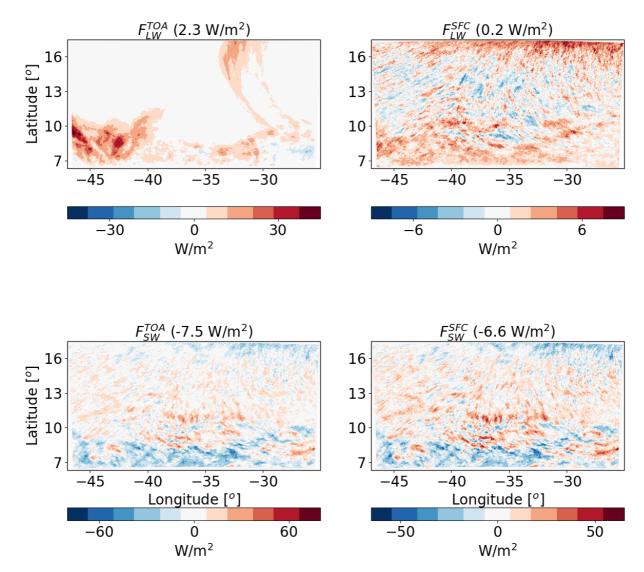




Figure 7. The differences between polluted (CDNC = 200 cm<sup>-3</sup>) and clean (CDNC = 20 cm<sup>-3</sup>) ICON simulations of the time mean radiative longwave (LW) and shortwave (SW) fluxes at the top of atmosphere (TOA) and surface (SFC) for the shallow-cloud dominated case (10-12/08/2016). The domain and time mean value of each term appears in parenthesis.

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The differences in the energy (Fig. 6) and radiation (Fig. 7) budgets between the clean and

polluted cases shown above, could be explained by the differences in the cloud mean properties.

Figure 8 presents the time evolution of some of the domain mean properties while Fig. 9 presents

time and horizontal mean vertical profiles. To examine the robustness of the trends we add here

two more CDNC cases of 100 and 500 cm<sup>-3</sup> (on top of the two that were examine above -20 and 315 200 cm<sup>-3</sup>). Figure 8 demonstrates that the domain mean cloud fraction (CF) generally decreases 316 with the increase in CDNC (except for the first ~10 hours of the simulations). Examining the 317 vertical structure of the CF response (Fig. 9), demonstrates that with the increase in CDNC there 318 is a reduction in the low level (below 800 mb) CF concomitantly with an increase in CF at the 319 middle and upper troposphere. The differences in rain rate between the different simulations are 320 small. However, both the liquid water path (LWP) and the ice water path (IWP) show a consistent 321 increase with CDNC. Accordingly, also the total water path (TWP), which is the sum of the LWP 322 323 and the IWP, substantial increases with CDNC. The vertical profiles of the different hydrometers (Fig. 9) indicate, as expected, that the cloud droplet mass mixing ration (qc - droplet with radius 324 smaller than 40 µm) increases with CDNC, while the rain mass mixing ratio (qr - drops with 325 radius larger than 40 µm) decreases due to the shift in the droplet size distribution to smaller 326 327 sizes under larger CDNC conditions. As this case is dominated by shallow clouds, there exists only a comparably small amount of ice mixing ration (qi) (c.f. Fig. 17), but its concentration 328 329 increases with the CDNC increase. The combined effect of the increase in CDNC is to monotonically increase the total water mixing ratio (qt) above 800 mb (Fig. 9). The relative 330 331 increase in qt with CDNC becomes larger at higher levels.

The increase in cloud water with increasing CDNC can explain both the reductions in the net 332 downward SW fluxes (both at TOA and surface) and the decrease in outgoing LW flux at TOA 333 (Fig. 7), as it results in more SW reflection concomitantly with more LW trapping in the 334 335 atmosphere (Koren et al., 2010). Another contributor to the SW flux reduction (more reflectance) at the TOA is the Twomey effect (Twomey, 1977), while, the decrease in the low-level CF 336 337 compensates some of this effect. Here we present the outcome of these contradicting effects on the SW fluxes, which shows a reduction at both the TOA and surface (Fig. 7). For estimating the 338 339 relative contribution of the Twomey effect compare to the cloud adjustments (CF and TWP 340 effects) to the SW flux changes, we have re-run the simulations with the Twomey effect turned 341 off (the radiation calculations do not consider the changes in effective radius between the different simulations). It demonstrates that without the Twomey effect the TOA SW difference 342 is only -1.7  $W/m^2$  as compared to -7.5  $W/m^2$  with the Twomey effect, demonstrating the 343 predominant role of the Twomey effect. For estimating the relative contribution of the changes 344 in CF and water content to the SW flux changes we have conducted off-line radiative transfer 345 sensitivity tests. To quantify the water content radiative effect, we feed the same CF vertical 346 profile from the model into the offline radiative transfer model BUGSrad, while allowing the 347

348 water content vertical profile to change (and visa versa to compute the CF radiative effect). This 349 approach demonstrates that the contribution from the small reduction in CF is negligible 350 compared to the increased SW reflectance caused by the increased water content (the effect of 351 the reduction in CF compensate only about 1% of the effect of the increase in the water content).

We also note a monotonic increase in the near surface temperature with CDNC (see also Fig. 10 352 below). This trend can be explained by warm rain suppression with increasing CDNC that leads 353 to less evaporative cooling (see the decrease in the total amount of water mass mixing ration just 354 above the surface in Fig. 9, (Dagan et al., 2016; Albrecht, 1993; Seigel, 2014; Seifert and Heus, 355 2013; Lebo and Morrison, 2014)). In addition, it was shown that under polluted conditions the 356 rain drops below cloud base are larger, hence evaporating less efficiently (Lebo and Morrison, 357 358 2014; Dagan et al., 2016). The increase in the near surface temperature drives the decrease in the 359  $Q_{SH}$  (Fig. 6).

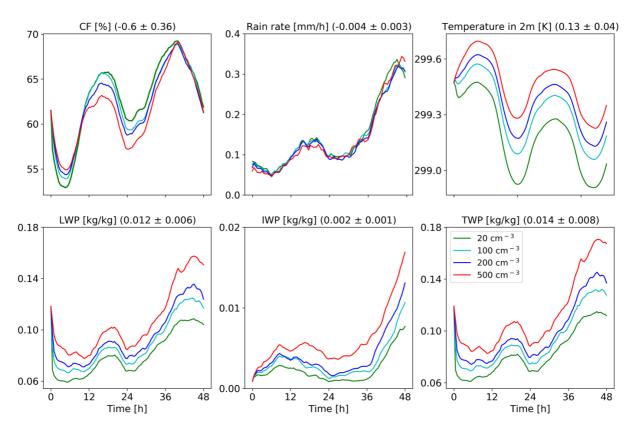
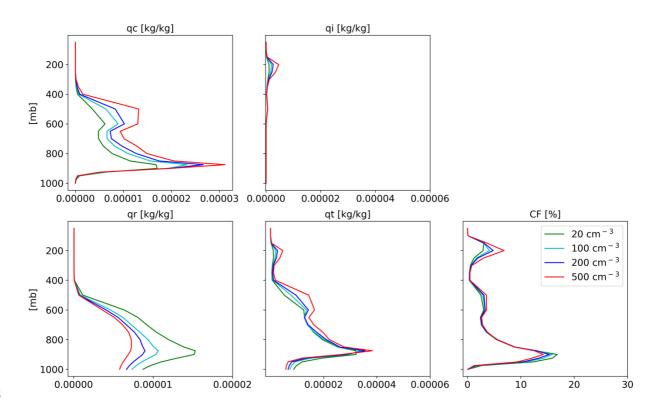




Figure 8. Domain average properties as a function of time for the different CDNC simulations for the shallowcloud dominated case. The properties that are presented here are: cloud fraction (CF), rain rate, temperature in 2 m, liquid water path (LWP – based on the cloud water mass, excluding the rain mass for consistency with satellite observations), ice water path (IWP) and total water path (TPW = LWP + IWP). For each property, the mean difference between all combinations of simulations, normalized to a factor 5 increase in CDNC, and its standard deviation appear in parenthesis.



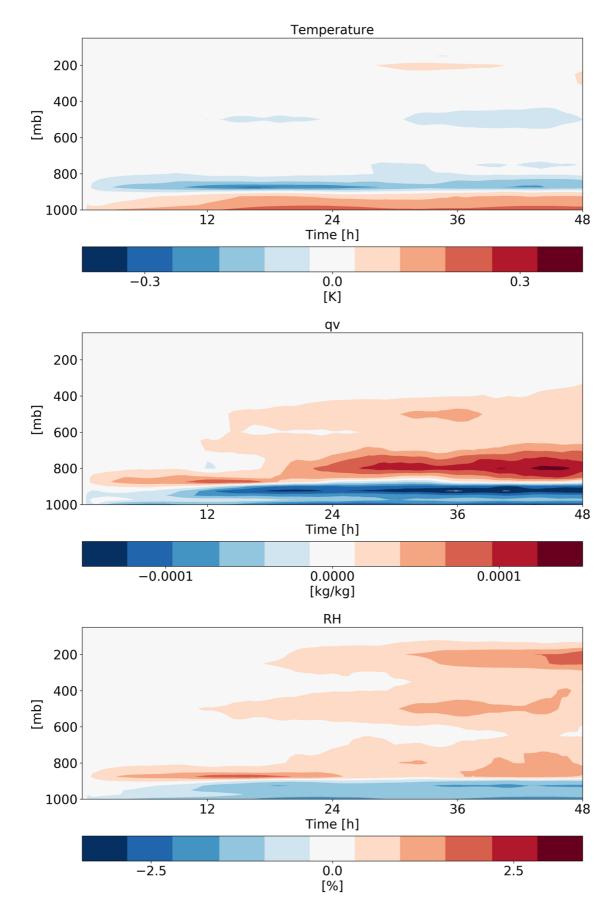
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Figure 9. Domain and time average vertical profiles for the different CDNC simulations for the shallow-cloud
dominated case. The properties that are presented here are: cloud droplet mass mixing ratio (qc – for clouds'
droplets with radius smaller than 40 µm), ice mass mixing ratio (qi), rain mass mixing ratio (qr - for clouds'
drops with radius larger than 40 µm), total water mass mixing ratio (qt = qc+qi+qr), and cloud fraction (CF).
The x-axis ranges are identical as for the deep-cloud dominated case – Fig. 17.

In addition to the clouds' effect on the radiation fluxes, changes in humidity could also contribute 375 (Fig. 10). We note that increase in CDNC leads to increase in relative humidity (RH) and specific 376 humidity (qv) at the middle and upper troposphere without a significant temperature change. The 377 increased humidity at the upper troposphere would act to decrease the outgoing LW flux, similar 378 to the effect of the increased ice content in the upper troposphere (Fig. 9). However, sensitivity 379 studies with off-line radiative transfer calculations using BUGSrad demonstrate that the vast 380 majority (more than 99%) of the different in  $F_{LW}^{TOA}$  between clean and polluted conditions emerges 381 from the cloudy skies (rather than clear-sky), suggesting that the effect of the increased ice 382 383 content at the upper troposphere dominates.

Both the increase in water vapor and ice content in the upper troposphere are driven by an increase in upward water (liquid and ice) mass flux with increasing CDNC (Fig. 11). An increase 386 in mass flux could be caused by an increase in vertical velocities and/or by an increase in cloud (or updraft) fraction and/or by an increase in cloud water content. In our case, the increases in 387 mass flux is driven partially by the small increase in vertical velocity (especially for updraft 388 between 5 and 10 m/s – Fig. 11), partially by the small increase in cloud faction at this level (Fig. 389 390 9) and mostly due to the larger water mass mixing ratio (Fig. 9) that leads to an increase in mass flux even for a given vertical velocity. The increased relative humidity at the upper troposphere, 391 392 further increases the ice particle lifetime at these levels (in addition to the microphysical effect (Grabowski and Morrison, 2016)) as the evaporation rate decreases. In addition, the differences 393 394 in the thermodynamics evolution between the different simulations (Fig. 10) demonstrate drying and warming of the boundary layer with increasing CDNC, due to reduction in rain evaporation 395 below cloud base and deepening of the boundary layer (Dagan et al., 2016; Lebo and Morrison, 396 2014; Seifert et al., 2015; Spill et al., 2019). The drying of the boundary layer could explain the 397

reduction in the low cloud fraction (Fig. 9 (Seifert et al., 2015)).



- 400 Figure 10. Time-height diagrams of the differences in the domain mean temperature, specific humidity (qv)
- 401 and relative humidity (RH) vertical profiles between polluted (CDNC =  $200 \text{ cm}^{-3}$ ) and clean (CDNC =  $20 \text{ cm}^{-3}$ )
- 402 <sup>3</sup>) simulations for the shallow-cloud dominated case (10-12/08/2016).
- 403

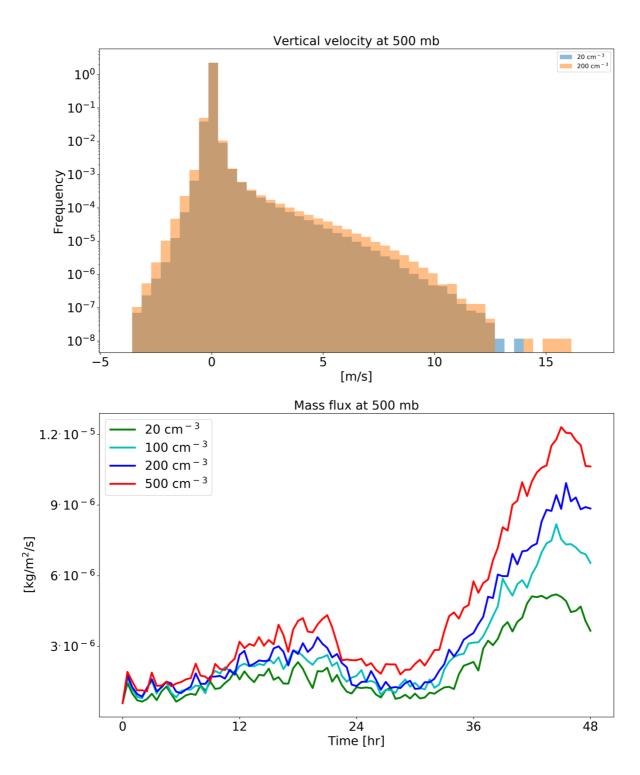


Figure 11. histograms of ICON simulated vertical velocity at the level of 500 mb for a clean (CDNC = 20 cm<sup>-3</sup>)
and polluted (CDNC = 200 cm<sup>-3</sup>) simulations (upper), and the time evolution of the net upwards water
(liquid and ice) mass flux (lower) for the different CDNC simulations for the shallow-cloud dominated case

408 (10-12/08/2016). The 500 mb level is chosen as it represents the transition between the warm part to the cold
 409 part of the clouds. In the histogram only two simulations are presented for clarity.

410

#### 411 Deep-cloud dominated case -16-18/08/2016

Next, we analyse the atmospheric energy budget for the deep-cloud dominated case (Fiona 412 tropical storm – Fig. 12). As opposed to the shallow-cloud dominated case, in this case the LP 413 contribution dominates over the radiative cooling and hence the energy imbalance R is positive 414 and large, suggesting divergence of dry static energy out of the domain. This difference in the 415 416 base line atmospheric energy budget between the different cases simulated here, enable an 417 examination of the aerosol effect on the atmospheric energy budget under contrasting initial conditions. As in the shallow-cloud dominated case, the  $Q_R$  values varies between small values 418 419 (especially at the regions that were mostly covered by deep clouds) to larger negative values (dominated at the regions that were coved by shallow clouds). The  $Q_{SH}$  is positive and an order 420 of magnitude smaller than the  $Q_R$  and LP, similar to the shallow-cloud dominated case. 421

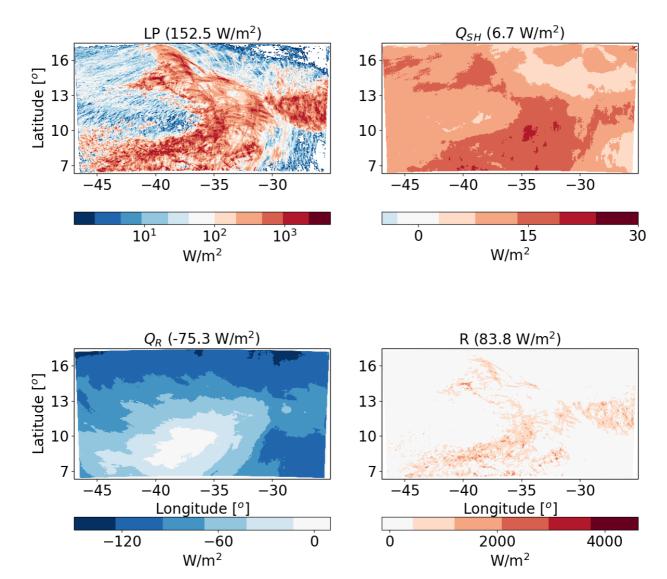
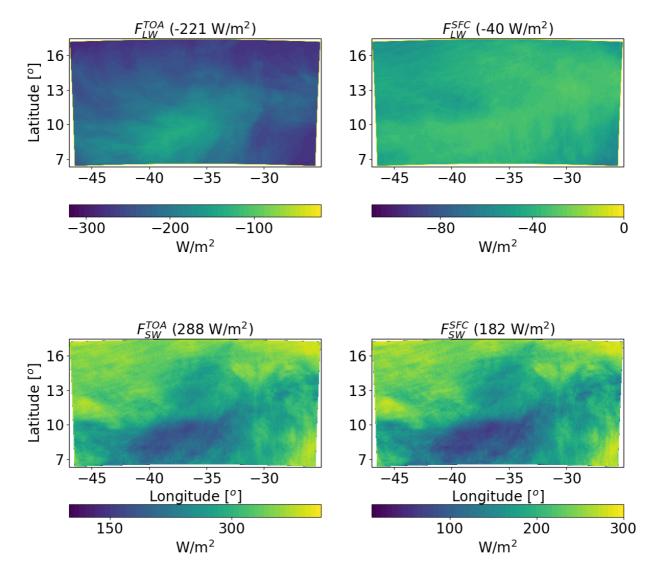




Figure 12. Spatial distribution of the time mean of the different terms of the energy budget for the ICON simulation of the deep-cloud dominated case (16-18/08/2016) with CDNC = 20 cm<sup>-3</sup>. The terms that appear here are: *LP* - latent heat by precipitation,  $Q_{SH}$  - sensible heat flux,  $Q_R$  - atmospheric radiative warming, and R – the energy imbalance. The domain and time-mean value of each term appears in parenthesis.

Further examination of the radiative fluxes (Fig. 13) demonstrates again the resemblance in the spatial structure between  $Q_R$  and  $F_{LW}^{TOA}$ . As compared to the shallow-cloud dominated case, since the clouds are more opaque and cover larger fraction of the sky, there is a decrease in the magnitude of all fluxes (in different amount). For example,  $F_{SW}^{SFC}$  is lower by 41 W/m<sup>2</sup> (representing larger SW reflectance back to space) and the magnitude of  $F_{LW}^{TOA}$  by 47 W/m<sup>2</sup> as compare to the shallow-cloud dominated case. The combined effect of the radiative flux

434 differences between the two cases is a decrease of the atmospheric radiative cooling by 39.6 435  $W/m^2$  (-114.7 compare with -75.3  $W/m^2$  – see Figs. 5 and 13).



436

Figure 13. Spatial distribution of ICON simulated time-mean longwave (LW) and shortwave (SW) radiation
fluxes at the top of atmosphere (TOA) and surface (SFC) for a simulation of the deep-cloud dominated case
(16-18/08/2016) with CDNC = 20 cm<sup>-3</sup>. The domain and time mean value of each term appears in parenthesis.

#### 441 <u>Response to aerosol perturbation – deep-cloud dominated case</u>

For the deep-cloud dominated case, an increase in CDNC results in a decrease in *LP* by -0.3 W/m<sup>2</sup>. Again, this difference is due to a non-statistically significant precipitation changes (see also Fig. 16 below). A similar  $Q_{SH}$  decrease as in the shallow-cloud dominated case is observed in the deep-clouds dominated case (see Figs. 14 and 6). The predominant difference in the response between the two cases is in  $Q_R$ , which increases much more in the deep-cloud dominated case: 10.0 W/m<sup>2</sup> (Fig. 14) compared with 1.6 W/m<sup>2</sup> in the shallow-cloud dominated case (Fig. 6).

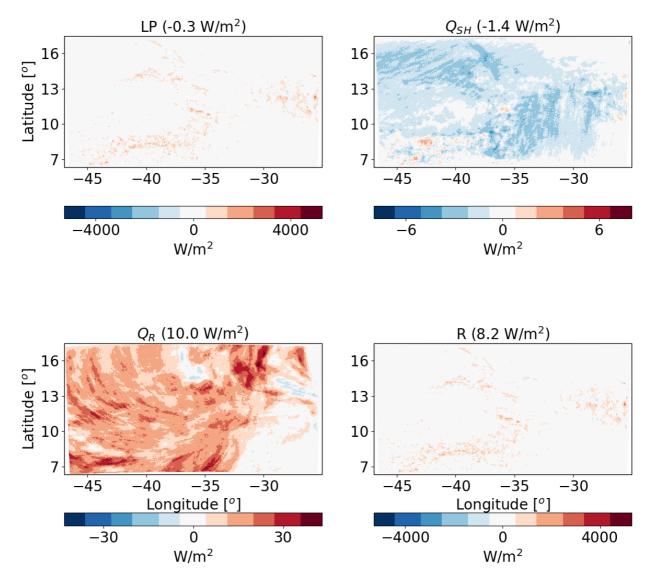
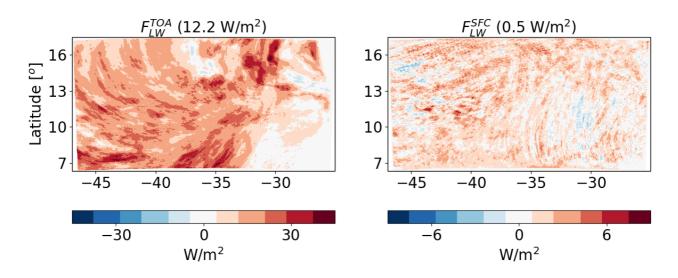


Figure 14. The differences between polluted (CDNC = 200 cm<sup>-3</sup>) and clean (CDNC = 20 cm<sup>-3</sup>) ICON simulations of the time-mean terms of the energy budget for the deep-cloud dominated case (16-18/08/2016). The terms that appears here are: LP - latent heat by precipitation,  $Q_{SH}$  - sensible heat flux,  $Q_R$  - atmospheric radiative warming, and R – the energy imbalance. The domain and time mean value of each term appears in parenthesis.

455

The large increase in  $Q_R$  is caused mostly by the increase in  $F_{LW}^{TOA}$  (which becomes less negative i.e. less outgoing LW radiation under polluted conditions – Fig. 15). The CDNC effect on  $F_{LW}^{SFC}$ has a much smaller magnitude. The SW fluxes changes are substantial (-14.1 W/m<sup>2</sup> at TOA and

-12.3 W/m<sup>2</sup> at the surface), however, in terms of the atmospheric energy budget, since clouds do
not absorb much in the SW, the TOA and surface changes almost cancel each other out and the
net effect is only ~1.8 W/m<sup>2</sup> atmospheric radiative cooling (which decrease some of the LW
warming). The net TOA total (SW+LW) radiative flux change is about -1.9 W/m<sup>2</sup>. The trends in
the mean cloud properties (Figs. 16 and 17 below) can explain this large radiative response.



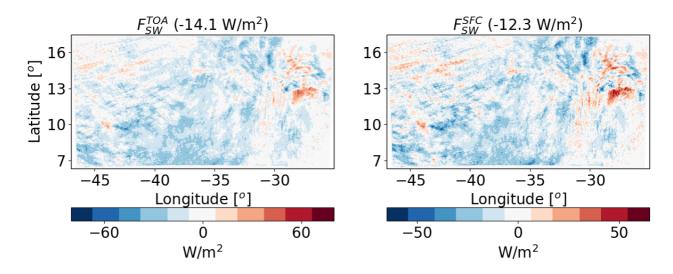


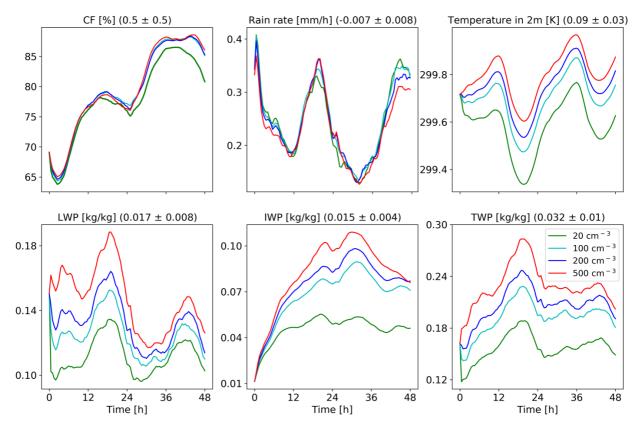


Figure 15. The differences between polluted (CDNC = 200 cm<sup>-3</sup>) and clean (CDNC = 20 cm<sup>-3</sup>) ICON simulations of the time mean radiative longwave (LW) and shortwave (SW) fluxes at the top of atmosphere (TOA) and surface (SFC) for the deep-cloud dominated case (16-18/08/2016). The domain and time mean value of each term appears in parenthesis.

470 Figure 16 presents some of the domain mean properties as a function of time for the deep-cloud dominated case. It demonstrates an increase in CF with CDNC which is more significant during 471 the second day of the simulation. This is opposite to the CF reduction in the shallow-cloud 472 dominated case (Fig. 8). It also demonstrates a very significant increase in LWP and, even more 473 474 (in relative terms), in IWP and thus also in TWP. The increase in CF and water content can explain the decrease in SW fluxes both at TOA and surface (Fig. 15) as more SW is being 475 reflected back to space. The larger SW reflection under increased CDNC is also contributed to 476 by the Twomey effect (Twomey, 1977). Re-running the simulations without the Twomey effect 477 result in 9.6 W/m<sup>2</sup> reduction in the TOA SW flux as compare to 14.1 W/m<sup>2</sup> with the Twomey 478 effect on. We note that the relative role of the Twomey effect (compare to the cloud adjustments 479 - CF and TWP) is larger in the shallow-cloud dominated case as compared to the deep-cloud 480 dominated case (-14.1 W/m<sup>2</sup> and -9.6 W/m<sup>2</sup> for simulations with and without the Twomey effect 481 in the deep-cloud dominated case, compare to  $-7.5 \text{ W/m}^2$  and  $-1.7 \text{ W/m}^2$  in the shallow-cloud 482 dominated case, respectively). However, it should be noted that the Twomey effect due to 483 changes in the ice particles size distribution was not considered. In this case, unlike in the 484 shallow-cloud dominated case, the three contributions to the SW changes (CF, Twomey and 485 LWP/IWP, e.g. (Goren and Rosenfeld, 2014)) all contribute to the SW flux reduction (Fig. 15 486 487 presents the results of all contributors). Off-line sensitivity tests demonstrate that the relative contribution of the water content and the CF to the increase in SW reflectance is roughly <sup>3</sup>/<sub>4</sub> and 488 <sup>1</sup>/<sub>4</sub>, respectively. 489

490 The vertical profile changes with CDNC (Fig. 17) demonstrate a consistent picture of a decrease in CF in low clouds and a significant increase in CF and liquid and ice content at the mid and 491 upper troposphere. The CF increase at the upper troposphere, and especially the increase in the 492 ice content, can explain the decrease in the outgoing LW radiation (Fig. 15). The increase in ice 493 content at the upper troposphere is in agreement with recent observational studies (Gryspeerdt et 494 495 al., 2018; Sourdeval et al., 2018; Christensen et al., 2016). Analysis of the upward water mass flux from the warm to the cold part of the clouds (at 500 mb) in the different simulations (Fig. 496 19), demonstrates a substantial increase with the increase in CDNC (Chen et al., 2017), which 497 occurs due to the increase in the water content (Fig. 17) and the delay in the rain formation to 498 higher levels (Heikenfeld et al., 2019), even without a large change in the vertical velocity or 499 cloud fraction at this level (Fig.17). Similar to the shallow-cloud dominated case (Fig. 8), the 500 near surface temperature monotonically increases with CDNC, while the effect on the mean rain 501 rate is small. 502

503 The differences in the thermodynamic evolution between polluted and clean conditions for this case (Fig. 18), demonstrate the same trend as in the shallow-cloud dominated case (Fig. 10). 504 Here again, we note an increase in the humidity at the mid and upper troposphere, that contribute 505 to the reduction in the outgoing LW flux. The deepening, drying and warming of the boundary 506 layer are observed in this case as well. Both the increase in humidity at the mid-upper troposphere 507 and the deepening of the boundary layer (Seifert et al., 2015) could cause a reduction of the 508 509 outgoing LW flux. To distinguished the effect of clouds and humidity at the different levels on the outgoing LW flux, we have conducted sensitivity off-line radiative transfer calculations using 510 511 BUGSrad. As in the shallow-cloud dominated case, the difference in outgoing LW flux between clean and polluted conditions primarily emerges from the CDNC effect on clouds. The small 512 remaining effect of the clear sky ( $\sim 0.2 \text{ W/m}^2$ ) is contributed by the change in the humidity at the 513 mid and upper troposphere rather than by the deepening of the boundary layer (which would lead 514 to LW emission from lower temperatures and is expected to be more significant under lower free 515 troposphere humidity conditions). 516



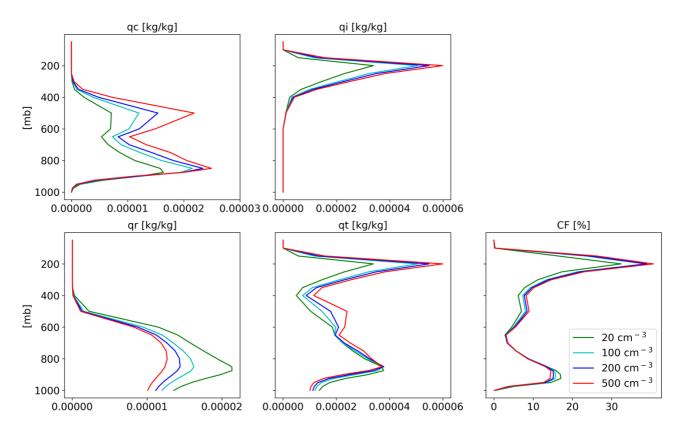


518 Figure 16. Domain average properties as a function of time for the different CDNC simulations for the deep-519 cloud dominated case. The properties that are presented here are: cloud fraction (CF), rain rate, temperature 520 in 2 m, liquid water path (LWP – based on the cloud water mass, excluding the rain mass for consistency 521 with satellite observations), ice water path (IWP) and total water path (TPW = LWP + IWP). For each

522 property, the mean difference between all combinations of simulations, normalized to a factor 5 increase in

523 CDNC, and its standard deviation appear in parenthesis.

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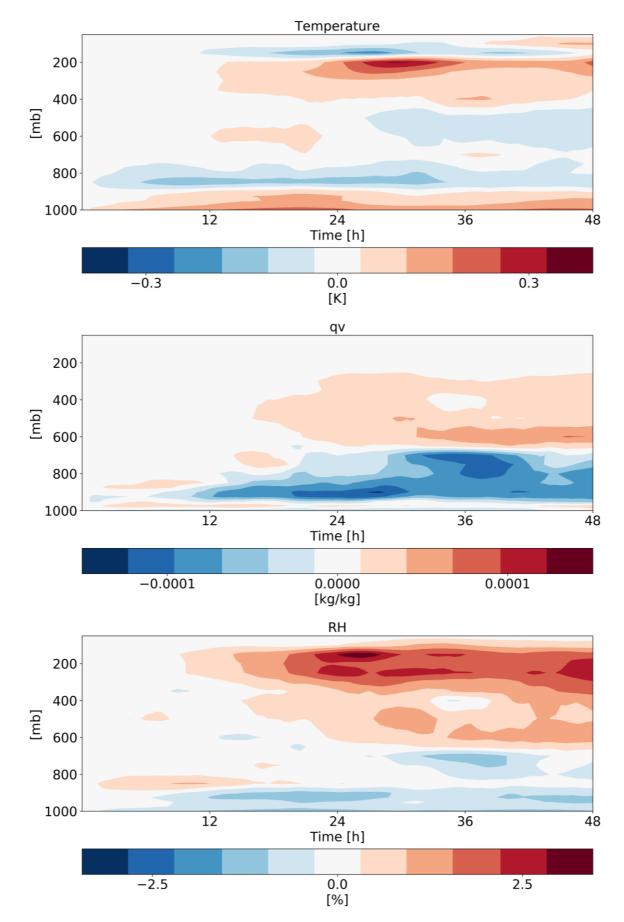


526 Figure 17. Domain and time average vertical profiles for the different CDNC simulations for the shallow-

527 cloud dominated case. The properties that are presented here are: cloud droplet mass mixing ratio (qc - for
 528 clouds' droplets with radius smaller than 40 μm), ice mass mixing ratio (qi), rain mass mixing ratio (qr - for

529 clouds' drops with radius larger than 40  $\mu$ m), total water mass mixing ratio (qt = qc+qi+qr), and cloud

530 fraction (CF).





532 Figure 18. Time-height diagrams of the differences in the domain mean temperature, specific humidity (qv)

and relative humidity (RH) vertical profiles between polluted (CDNC = 200 cm<sup>-3</sup>) and clean (CDNC = 20 cm<sup>-3</sup>)

534 <sup>3</sup>) simulations for the deep-cloud dominated case (16-18/08/2016).

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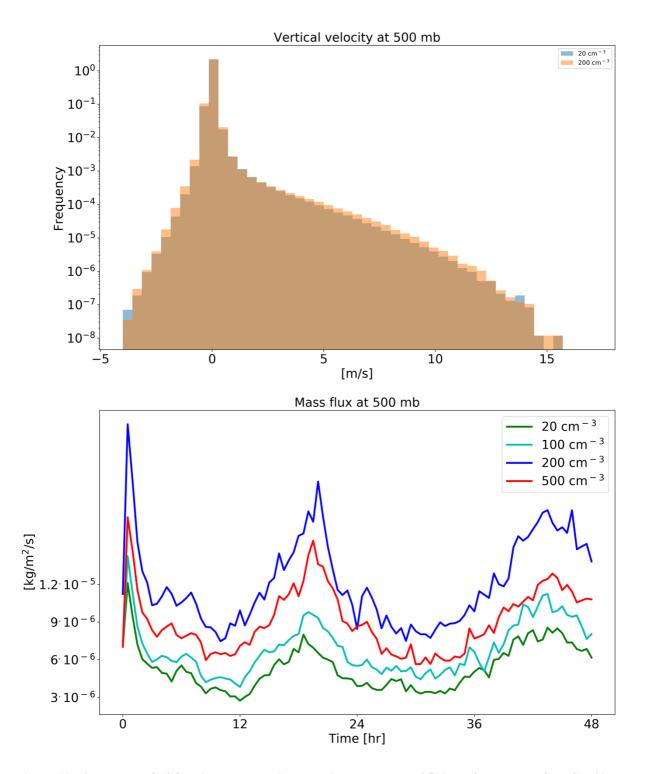


Figure 19. histograms of ICON simulated vertical velocity at the level of 500 mb for a clean (CDNC = 20 cm<sup>-</sup>
<sup>3</sup>) and polluted (CDNC = 200 cm<sup>-3</sup>) simulations (upper), and the time evolution of the net upwards water
(liquid and ice) mass flux (lower) for the different CDNC simulations for the deep-cloud dominated case (16-

540 18/08/2016). The 500 mb level is chosen as it represents the transition between the warm part to the cold part
541 of the clouds. In the histogram only two simulations are presented for clarity.

542

## 543 Summary and conclusions

Two different case studies of tropical cloud systems over the Atlantic Ocean were simulated 544 using the ICON numerical model in a cloud resolving configuration with 1.2 km resolution and 545 a relatively large domain (~22° x 11°). The cases represent dates from the NARVAL 2 field 546 campaign that took place during August 2016 and have different dominant cloud types and 547 different dominating terms in their energy budget. The first case (10-12/8/2016) is shallow-cloud 548 dominated and hence dominated by radiative cooling, while the second case (16-18/8/2016) is 549 dominated by deep convective clouds and hence dominated by precipitation warming. The main 550 objective of this study is to analyse the response of the atmospheric energy budget to changes in 551 cloud droplet number concentration (CDNC), which serve as a proxy for (or idealized 552 representation of) changes in aerosol concentration. This enables better understanding of the 553 554 processes acting in global-scale studies trying to constrain aerosol effect on precipitation changes using the energy budget perspective (O'Gorman et al., 2012; Muller and O'Gorman, 2011; 555 556 Hodnebrog et al., 2016; Samset et al., 2016; Myhre et al., 2017; Liu et al., 2018; Richardson et al., 2018; Dagan et al., 2019a). Our results demonstrate that regional atmospheric energy budgets 557 558 can be significantly perturbed by changes in CDNC and that the magnitude of the effect is cloud 559 regime dependent (even for a given geographical region and given time of the year as the two cases are separated by less than a week). 560

Figure 20 summarizes the energy and radiation response of the two simulated cases to CDNC 561 perturbations. It shows that the atmosphere in the deep-cloud dominated case experiences a very 562 strong atmospheric warming due to an increase in CDNC ( $10.0 \text{ W/m}^2$ ). Most of this warming is 563 caused by a reduction in the outgoing LW radiation at the TOA. The SW radiative fluxes (both 564 at the TOA and surface) is also significantly modified but their net effect on the atmospheric 565 column energy budget is small. The net TOA radiative fluxes change in this case is -1.9 W/m<sup>2</sup>. 566 Beside the atmospheric radiative warming, changes in precipitation (~-0.3 W/m<sup>2</sup>), and in sensible 567 heat flux ( $Q_{SH}$ , -1.4 W/m<sup>2</sup>) also contribute to the total trend as a response of increase in CDNC. 568 We note that since 1 mm/hr of rain is equivalent to 628 W/m<sup>2</sup>, even negligible changes in 569 precipitation of less than 0.5 mm over 48 hr (as seen in our simulations) can still appear as 570 significant changes in the atmospheric energy budget and contribute a few W/m<sup>2</sup>. 571

The response of the radiative fluxes can be explained by the changes in the mean cloud and 572 thermodynamic properties in the domain. The mean cloud fraction (CF) increases with the 573 increase in CDNC (Fig. 16) while the vertical structure of it indicates a reduction in the low 574 cloud fraction (below 800 mb) and an increase in the mid and upper troposphere CF (Fig. 17). 575 The water content (both liquid and ice) also increase with the increase in CDNC (Figs. 16 and 576 17) with increasing amount with height. These changes in the mean cloud properties drive both 577 the reduction in SW fluxes at TOA and surface and LW flux at TOA as the clouds become more 578 opaque (Koren et al., 2010; Storelvmo et al., 2011) and cover a larger fraction of the sky. In 579 580 addition to cloud responses, the domain-mean thermodynamic conditions change as well (Fig. 18). Specifically, the humidity content at the mid and upper troposphere increases with higher 581 CDNC, (due to increase mass flux to the upper troposphere) which further decreases the outgoing 582 LW flux at the TOA. However, the vast majority of the LW effect emerges from the changes in 583 584 clouds.

Both the increase in water vapor and ice content in the upper troposphere are driven by an 585 586 increase in water mass flux with increasing CDNC to these levels (Fig. 19, (Koren et al., 2005; Rosenfeld et al., 2008; Altaratz et al., 2014; Chen et al., 2017)), which is caused mostly by the 587 588 increase in the water mixing ratio in the mid-troposphere rather than by increase in vertical velocity (Fig. 19) or in cloud fraction (Fig. 17). The ice content in the upper troposphere is also 589 increased due to reduction in the ice falling speed (Grabowski and Morrison, 2016), while the 590 increased relative humidity at these levels, further increases the ice particle lifetime due to slower 591 evaporation. However, the increase in water mass flux to the upper layers is not accompanied 592 with an increase in precipitation as predicted by the classical "invigoration" paradigm (Altaratz 593 et al., 2014; Rosenfeld et al., 2008), which suggest that some compensating mechanisms are 594 operating (Stevens and Feingold, 2009). 595

In the shallow-cloud dominated case (which also contains a significant amount of deep 596 convection), the response of  $Q_R$  is weaker but still substantial (a total decrease in the atmospheric 597 radiative cooling of 1.6  $W/m^2$  - Fig. 20). The weaker total response under the shallow-cloud 598 dominated conditions is due to the smaller role of the ice part in this case. Here again, the changes 599 in  $Q_{SH}$  decrease about -1.4 W/m<sup>2</sup> of this atmospheric warming. As in the deep-cloud dominated 600 case, most of the atmospheric radiative warming is caused by reduction in the outgoing LW flux, 601 while the surface and TOA SW fluxes changes are non-negligible but cancel each other out (in 602 terms of the atmospheric energy budget – reflecting small SW atmospheric absorption changes). 603 604 However, a significant TOA net (SW+LW) radiative flux change of ~-5.2 W/m<sup>2</sup> remains. In this 605 case, the cloud-mean effect on radiation is more complicated. While CF decreases with increasing CDNC, the mean water path (both LWP and IWP) increases (Fig. 8). As in the deep-606 cloud dominated case, the increase in the water content occurs mostly at the mid and upper 607 troposphere, while the decrease in CF occurs mostly in the lower troposphere (Fig. 9). In terms 608 609 of the SW fluxes, the effect of the decrease in low CF (decrease SW reflections) and the increase 610 in water mass (increase SW reflections) would partially compensate, while the Twomey effect (Twomey, 1977) adds to the increase SW reflections. In this case, the net effect is more SW 611 reflected back to space at TOA and a net negative flux change (including also the LW). 612

613 There exists a large spread in estimates of aerosol effects on clouds for different cloud types and 614 different environmental conditions. In this study, as we use a relatively large domain  $(22^{\circ} \times 11^{\circ})$ 615 and two different dates (each for two days), we sample many different local environmental conditions and cloud types. Such more realistic setups (although with lower spatial resolution) 616 617 could provide more reliable estimates of aerosol effects on heterogeneous cloud systems than just one-cloud-type, small domain simulations (as was done in many previous studies, e.g (Dagan 618 619 et al., 2017; Seifert et al., 2015; Ovchinnikov et al., 2014)). However, the conclusions demonstrated here are based on two specific cases. In order to examine the validity of our main 620 621 conclusions over a wider range of initial conditions, we have conducted a large ensemble of 622 simulations starting from realistic initial conditions (although with a smaller domain) in a companion paper (Dagan and Stier, 2019). These simulations demonstrate that the main 623 conclusions presented in this paper are robust and hold also for a wide range of initial conditions 624 representative for this area. In addition, the realistic setup with the continuously changing 625 boundary conditions and systems that pass through the domain, which are used here, prevent 626 conclusions that might be valid only in cyclic double periodic large eddy simulations, as the 627 background meteorological conditions change more realistically (Dagan et al., 2018b). Another 628 629 uncertainty in the assessment of the aerosol response are the large differences between different 630 models and microphysical schemes (White et al., 2017; Fan et al., 2016; Khain et al., 2015; Heikenfeld et al., 2019). In this study, as we use only one model, we do not address this 631 632 uncertainty. In future work we intend to examine the response in multiple models. In addition, more detailed observational constraints on the models are needed. Furthermore, we do not 633 include the temporal evolution of the aerosol concentration. Feedbacks between the aerosol 634 concentration and clouds processes (such as wet scavenging), as well as the direct effects of 635 aerosol on radiation would add another layer of complexity that should be accounted for in future 636 637 work.

638 Generally, the global mean aerosol radiative forcing is estimated to be negative (Boucher et al., 2013; Bellouin et al., 2019). However, these global aerosol forcing estimates have so far not 639 included the radiative forcing associated with potential effects of aerosols on deep convection -640 and these effects are not represented in most current climate models due to limitations in 641 convection parameterisations, with only a few exceptions (Kipling et al., 2017; Labbouz et al., 642 2018). Here we demonstrate the existence of non-negligible aerosol radiative effects (of -5.2 and 643 -1.9 W/m<sup>2</sup> for the shallow and deep cloud dominated cases, respectively) in tropical cloud 644 systems, that contained both deep and shallow convective clouds, with significant SW and LW 645 646 contributions. From the (limited) two cases simulated here, it appears that (in agreement with previous studies) the aerosol effect may be regime dependent and that even within a given cloud 647 regime the effect may vary with the meteorological conditions. 648

Finally, we hypothesise that the aerosol impact shown on the atmospheric energy balance, withincreasing divergence of dry static energy from deep convective regions concomitantly with

651 increased convergence in shallow clouds regions, can have effects on the large-scale circulation.

652 This should be investigated in future work.

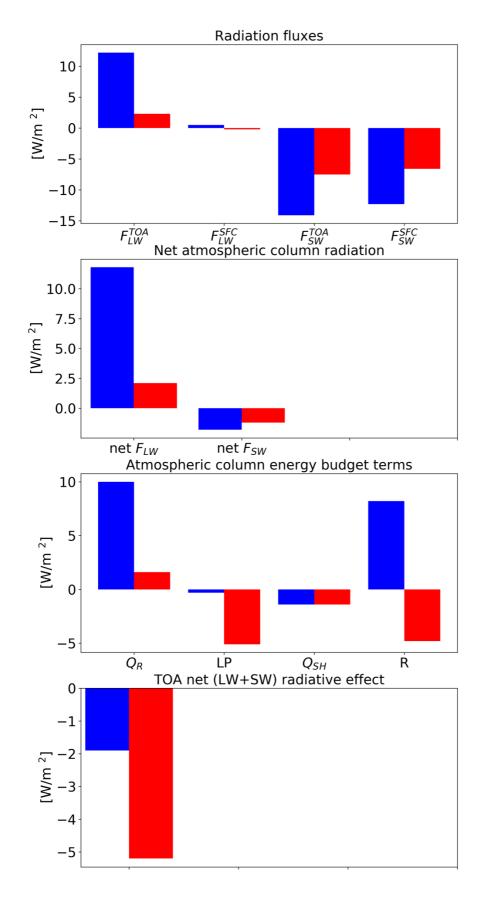




Figure 20. Summary of the radiation and energy response to CDNC perturbation in the two different cases.

Author contributions. G. D. carried out the simulations and analyses presented. G.C., D.K. and A.S. assisted with the simulations. M.C. assisted with the radiative transfer calculations and comparison with observations. P. S. and A.S. assisted with the design and interpretation of the analyses. G. D. prepared the manuscript with contributions from all co-authors.

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