1 Response to the reviewers' comments on

2 Atmospheric energy budget response to idealized aerosol perturbation in tropical cloud

3 <u>systems</u>

4 We would like to thank the revisers for their constructive and thoughtful reviews that helped

5 us improve our paper.

6 Below please find a point by point reply to all of the reviewers' comments (in blue).

7

8 **Reviewer #1:**

9 The authors have run some fairly large-domain cloud resolving simulations over the tropical

10 Atlantic, in order to investigate how aerosol indirect effects contribute to the radiative impact

11 of aerosols. They note a non-negligible contribution that depends on the cloud regime being

- 12 examined. The regime and location dependence of aerosol forcing is something that is not well
- 13 captured in climate models, since most don't include aerosol effects on convection. This study

14 is straightforward and worthwhile, as it begins to break down these differences in the radiation

budgets. I believe it is a good contribution and have only a few suggestions to help improve

16 the manuscript.

17 <u>Reply:</u> we would like to thank the reviewer again for the effort and the constructive comments.

- 18 We are happy that the reviewer found our paper to be straightforward and worthwhile.
- 19

20 Comments:

Changing CDNC is, you even admit, a rather simplistic way to approach aerosol effects. In addition to neglecting the activation and scavenging effects that you mention, another missing piece is the direct effect. This can be especially important in the eastern Atlantic where you are looking since a lot of the aerosols that would be present in this region are dust. Have you

25 considered how the direct effect would fit into this?

Reply: Thank you for this comment. Indeed, the direct effect of aerosol is interesting and 26 important [and is a main focus of our work, i.e. (Dagan et al., 2019)] but is not included in this 27 study. We believe that separating the overall response of the atmospheric energy budget to the 28 29 radiative and microphysical aerosol effects is a necessary first step in studying this complex system. This is the approach we are taking in the current study. However, we are currently 30 working on implementing the aerosol model HAM (Stier et al., 2005) into the reginal version 31 of ICON. This will allow studying the mutual interaction between the aerosol radiative and 32 33 microphysical effects in a cloud and aerosol resolving simulations.

34 Following this comment, we have added to the revised manuscript the following:

"The different CDNC scenarios serve as a proxy for different aerosol conditions (as the first 35 order effect of increased aerosol concentration on clouds is to increase the CDNC, Andreae, 36 2009). This also allows to separate the cloud response from the uncertainties involved in the 37 representation of the aerosols in numerical models (Ghan et al., 2011; Simpson et al., 2014; 38 39 Rothenberg et al., 2018). However, it limits potential feedbacks between clouds and aerosols, such as the removal of aerosol levels by precipitation scavenging and potential aerosol effects 40 thereon. In addition, the fixed CDNC framework does not capture the differences in aerosol 41 42 activation between shallow and deep clouds, due to differences in vertical velocity. Another aerosol effect that is not included in our simulations is the direct interaction between aerosol 43 and radiation. In future work we plan to examine the mutual interaction between microphysical 44 effects and the direct aerosol radiative effects." 45

46

47 In the conclusions:

48 *"Furthermore, we do not include the temporal evolution of the aerosol concentration.*

49 *Feedbacks between the aerosol concentration and clouds processes (such as wet scavenging),*

so as well as the direct effects of aerosol on radiation would add another layer of complexity that

- 51 *should be accounted for in future work.*"
- 52

You held SST constant in these simulations. Do you have a sense for how much this might
have affected the overall energy budgets? I would suspect at least the sensible heat flux might
show some differences.

Reply: We agree that including interactive SST would predominantly affect the sensible heat 56 flux. However, please note that in both cases the sensible heat flux is an order of magnitude 57 smaller than the rest of the terms (Figs. 4 and 12 in the manuscript). Hence, we do not believe 58 that the effect on the total energy budget would be large. In addition, due to the large heat 59 capacity of the ocean, over two days of simulation the SST is not expected to dramatically 60 change. For example, in the deep-cloud dominated case, the difference in the surface radiative 61 fluxes between the clean and polluted conditions is about 12 W/m^2 . Considering the heat 62 capacity of 50m deep ocean mixed layer results in about 0.005K difference in the SST between 63 the two cases over the two days simulation. In the shallow-cloud dominated case the difference 64 in surface radiative fluxes in about half of the deep-cloud dominated case (6 W/m²), resulting 65 in half the temperature change. 66

67 Following this argument, we added a clarification about this point to the revised manuscript:

68 "Additional details, such as the surface and atmospheric physics parameterizations, are 69 described in Klocke et al., (2017) and include an interactive surface flux scheme and fixed sea 70 surface temperature (SST). We note that using a fixed SST does not include feedbacks of 71 aerosols on the SST evolution that could change the surface fluxes. However, due to the large 72 heat capacity of the ocean, we do not expect the SST to dramatically change over the two days 73 simulations."

74

Your 'residual' term is rather large, especially in the deep convective case. You make the point earlier that this term would become negligible on longer time and spatial scales, however an important point in this paper is how large the differences can be on smaller scales. Do you have thoughts on what is largely making up this residual term? How much of it is physical processes that you are not considering, versus the fact that the model simulations are not going to be perfectly balanced, considering the scales and the boundary forcing.

<u>Reply:</u> What was previously called a "residual" term was changes in the revised manuscript to
be refer to as "energy imbalance" as it better describes it. A recent study shows that in order to
get close to energy balance the spatial scale should be on the order of ~5000km (Jakob et al.,
2019) and the time scale longer than a month (we found a similar scale using GCMs). Our
simulations operate on smaller spatial-temporal scales than that and hence it is not surprising
that we obtain an imbalance.

The energy imbalance is composed of changes in the storage term and local divergence or convergence of dry-static energy into the domain. In our case, almost the entire imbalance is simply dry static energy that moves in or out of the domain. For example, in the deep-cloud dominated case there is a net production of dry-static energy in the domain by precipitation (which is not entirely balanced by the radiative cooling). This extra dry-static energy is then advected out of the domain.

93 An explanation about this point was added to the revised manuscript:

94 *"The total column atmospheric energy budget can be described as follows:*

95 $LP + Q_R + Q_{SH} = div(s) + ds/dt$ (1)

96 Equation 1 presents a balance between the latent heating rate (LP - latent heat of condensation

97 [L] times the surface precipitation rate [P]), the surface sensible heat flux (Q_{SH}), the atmospheric

98 radiative heating (Q_R) , the divergence of dry static energy (div(s), which will become negligible

99 on sufficiently large spatial scales), and the dry static energy storage term (ds/dt, which will

100 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)."

103

"In this shallow-cloud dominated case the radiative cooling of the atmosphere is significantly
larger than the warming due to precipitation (mean of -114.7 W/m² compared to 90.1 W/m²),
hence the energy imbalance (R) is negative. Negative R means that there must be some
convergence of dry static energy into the domain and/or decrease in the storage term, in this
case it is mostly due to convergence of dry static energy."

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"Next, we analyse the atmospheric energy budget for the deep-cloud dominated case (Fiona tropical storm – Fig. 12). As opposed to the shallow-cloud dominated case, in this case the LP contribution dominates over the radiative cooling and hence the energy imbalance R is positive and large, suggesting divergence of dry static energy out of the domain."

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Your mass flux in Fig 11 - how is this calculated? Is this just a total over the whole domain?
Or only in updrafts? If this is domain-wide, I imagine the largest reason for the increase is
simply the larger amount of deep convection.

<u>Reply:</u> Thank you for your comment that helped us clarify this point. Calculating the relative 119 change in the cloud fraction and in the water content between clean and polluted conditions 120 demonstrate the dominate role of the latter in the increase in mass flux. This calculation 121 demonstrates that the cloud fraction (total water content) increases by 20% (72%) at 500mb in 122 the simulation with CDNC=200cm⁻³ compared with the simulations with CDNC=20cm⁻³ in the 123 124 deep-convection dominated case. Similarity, in the shallow dominated case the cloud fraction (total water content) increases by 22% (85%) at 500mb in the simulation with CDNC=200cm⁻³ 125 126 compared with the simulations with CDNC=20cm⁻³. These calculations demonstrate that about 80% of the increase in mass flux under polluted conditions occur due to the increase in water 127 content and only about 20% occur due to the increase in cloud fraction (recalling that the 128 vertical velocity is similar between the two simulations). The increase in total water content is 129 130 caused by warm rain suppression at the lower troposphere.

131 This is now better explained in the revised manuscript:

132 *"Both the increase in water vapor and ice content in the upper troposphere are driven by an*

133 increase in upward water (liquid and ice) mass flux with increasing CDNC (Fig. 11). An

increase 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 mass flux is driven partially by the small increase in vertical velocity (especially
for updraft between 5 and 10 m/s – Fig. 11), partially by the small increase in cloud faction at
this level (Fig. 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."

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"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. 19), demonstrates a substantial increase with the increase
in CDNC (Chen et al., 2017), which occurs due to the increase in the water content (Fig. 17)
and the delay in the rain formation to higher levels (Heikenfeld et al., 2019), even without a
large change in the vertical velocity or cloud fraction at this level (Fig. 17)."

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- 147

The paper is a bit long and I think you could consider getting rid of a few of the figures that 148 tell a redundant story. The inclusion of the 2nd, deep convective case is important because of 149 this point you make on page 32 "Our results demonstrate that regional atmospheric energy 150 151 budgets can be significantly perturbed by changes in CDNC and that the magnitude of the effect is cloud regime dependent (even for a given geographical region and given time of the 152 year as the two cases are separated by less than a week)." However, the physical mechanisms 153 for the changes in cloud amount and radiative fluxes are consistent between the two cases, so 154 155 some of the figures and discussion here are a bit repetitive.

156 <u>Reply:</u> Thank you for this comment. We did consider shortening the paper and not including 157 the figures of the second case but eventually we decided to keep them in as it demonstrate an 158 important point of this paper that the aerosol effect on the atmospheric energy budget is 159 meteorological conditions dependent.

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In general this could use some copy-editing. Nothing that prevents understanding, but there area number of small typos and verb agreement issues.

163 <u>Reply:</u> Thank you. The manuscript went thought copy editing and corrected accordingly.

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168 **Reviewer #2:**

- 169 This paper studies the differences in the radiative and energy budgets in a tropical environment
- 170 when the cloud droplet number concentration (CDNC) is changed (as a proxy for changes to
- the environmental aerosol concentration). The simulated periods are two separate 2-day within
- the same week where the convection is either predominantly shallow or predominantly deep -
- 173 each case is simulated with CDNC values. The authors find substantial changes in amount of
- 174 energy absorbed by the atmosphere by changing the CDNC and find substantial differences in
- the response between the two sets of simulations.
- 176 The main component of the paper is a breakdown of the energy budget into radiative, sensible
- 177 heating and heating through precipitation formation. The radiative component is later broken
- 178 down into shortwave and longwave fluxes at both the surface and top of the atmosphere. The
- 179 differences are also quantified in the time evolution of near- surface temperature, precipitation,
- 180 cloud fraction and in-cloud water contents.
- 181 Overall, I find the study to be well formulated with a clear motivation and simple but successful
- 182 strategy for breaking apart the components contributing to the changes in the atmospheric
- 183 energy budget. There are no substantial shortcomings that should prevent the publication of
- this study; however, I have a few suggestions that could improve this contribution which are
- 185 explained below.
- <u>Reply:</u> we would like to thank the reviewer again for the effort and for the suggestions. We are
 happy that the reviewer found that our paper well formulated.
- 188

189 Primarily my suggestions are aimed to help the authors achieve their stated aim of better understanding the physical processes behind aerosol effects on the atmospheric energy budget. 190 191 I see that their study does indeed achieve this, at least partly, but that these results are not clearly expressed in the abstract nor the conclusions. Throughout the paper, the authors do a 192 193 good job of describing the differences between their simulations and quantifying these differences (although in parts the quantification could be improved) - however, it is mostly left 194 to the reader to put these pieces of information together to get an understanding of the physical 195 processes involved. As a result, my overall impression of the authors conclusions and abstract 196 are: "we found another case where aerosol-cloud interactions behave differently under different 197 environmental conditions," which could be relatively simply converted to "these processes 198 (*see below) contribute to the different energy budget changes for shallow and deep convection 199 when CDNC is changed" 200

(1) From my understanding of the presented results, it seems that the large difference between 201 the shallow case and the deep case is the potential for a large upper-level cloud fraction change 202 in the deep case. I understand this as an increase of the anvil area, resulting in reduced LW 203 emission from the surface/lower atmosphere and therefore a warming contribution of the larger 204 anvil. In the shallow case, the upper level cloud fraction also has a systematic change, but 205 because it occupies a smaller part of the model domain - the overall change in the energy budget 206 is controlled by the change in low cloud fraction and the Twomey effect. If the authors agree 207 with this, I suggest adding a paragraph into the conclusions and a sentence in the abstract 208 209 clarifying these physical changes in the model and their impact on the energy budget.

<u>Reply:</u> Thank you for this suggestion that help us clarify this point. The reviewer's point is a
main conclusion of our paper; however, we believe that our results present more than just that
and include (among other) the effect of the thermodynamic evolution on radiation under
different CDNC conditions, the effect of CDNC on surface fluxes and so on. Nevertheless,
following the reviewer's comment, we have added a clarification to the revised manuscript.

In the abstract:

²¹⁶ "It is shown that the total column atmospheric radiative cooling is substantially reduced with ²¹⁷ CDNC in the deep-cloud dominated case (by $\sim 10.0 \text{ W/m}^2$), while a much smaller reduction ²¹⁸ ($\sim 1.6 \text{ W/m}^2$) is shown in the shallow-cloud dominated case. This trend is caused by an increase ²¹⁹ in the ice and water vapor content at the upper troposphere that leads to a reduced outgoing ²²⁰ longwave radiation, an effect which is stronger under deep-cloud dominated conditions."

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In the conclusions section:

"Both the increase in water vapor and ice content in the upper troposphere are driven by an 223 increase in water mass flux with increasing CDNC to these levels (Fig. 19, (Koren et al., 2005; 224 Rosenfeld et al., 2008; Altaratz et al., 2014; Chen et al., 2017)), which is caused mostly by the 225 increase in the water mixing ratio in the mid-troposphere rather than by increase in vertical 226 velocity (Fig. 19) or in cloud fraction (Fig. 17). The ice content in the upper troposphere is 227 also increased due to reduction in the ice falling speed (Grabowski and Morrison, 2016), while 228 the increased relative humidity at these levels, further increases the ice particle lifetime due to 229 slower evaporation." 230 "In the shallow-cloud dominated case (which also contains a significant amount of deep 231

231 In the shallow cloud dominated case (which also contains a significant amount of acception), the response of Q_R is weaker but still substantial (a total decrease in the 233 atmospheric radiative cooling of 1.6 W/m² - Fig. 20). The weaker total response under the

shallow-cloud dominated conditions is due to the smaller role of the ice part in this case."

236 (2) Breakdown of the vertical mass flux changes with CDNC into component parts

The vertical mass flux of water is shown to change between the simulations with different CDCN. What is the cause for this change? Either the vertical velocity should be increasing (which seems not to be the case from the vertical velocity distributions in Figures 11 and 19), so either the updraft area is increasing [implying wider updrafts?] or the in-cloud water mass in increasing [because of a less efficient precipitation-forming processes?]. To what extent are these two factors important? Furthermore, what happens to the vertical mass flux at (e.g.) 800 hPa - where the total water content is quite similar between all CDNC concentrations - is there

still an increased vertical mass flux?

<u>Reply:</u> Thank you for this comment that helped us clarify this point. Calculating the relative 245 change in the cloud fraction and in the water content between clean and polluted conditions 246 demonstrate the dominate role of the later in the increase in mass flux. This calculation 247 248 demonstrates that the cloud fraction (total water content) increases by 20% (72%) at 500mb in the simulation with CDNC=200cm⁻³ compared with the simulations with CDNC=20cm⁻³ in the 249 250 deep convection dominated case. Similarity, in the shallow dominated case the cloud fraction (total water content) increases by 22% (85%) at 500mb in the simulation with CDNC=200cm⁻³ 251 252 compared with the simulations with CDNC=20cm⁻³. These calculations demonstrate that about 80% of the increase in mass flux under polluted conditions occur due to the increase in water 253 254 content and only about 20% occur due to the increase in cloud fraction (recalling that the vertical velocity is similar between the two simulations). The increase in total water content is 255 caused by warm rain suppuration at the lower troposphere. 256

257 This is now better explained in the revised manuscript:

"Both the increase in water vapor and ice content in the upper troposphere are driven by an 258 increase in upward water (liquid and ice) mass flux with increasing CDNC (Fig. 11). An 259 260 increase 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 261 262 increases in mass flux is driven partially by the small increase in vertical velocity (especially for updraft between 5 and 10 m/s – Fig. 11), partially by the small increase in cloud faction at 263 this level (Fig. 9) and mostly due to the larger water mass mixing ratio (Fig. 9) that leads to 264 an increase in mass flux even for a given vertical velocity." 265

266

267 "Analysis of the upward water mass flux from the warm to the cold part of the clouds (at 500
268 mb) in the different simulations (Fig. 19), demonstrates a substantial increase with the increase

- in CDNC (Chen et al., 2017), which occur due to the increase in the water content (Fig. 17)
 and the delay in the rain formation to higher levels (Heikenfeld et al., 2019), even without a
- 271 *large change in the vertical velocity or cloud fraction at this level (Fig.17).*"
- 272
- In addition, we calculated the mass flux at the level of 800 mb as the reviewer suggested (Figs. R1 and R2 below). It demonstrates that in this level there is also a general increase in mass flux with the increase in CDNC but to a lesser extent. The increase in mass flux is driven by a small increase in water content (Figs. 9 and 17) and a small increase in vertical velocity (Figs. R1 and R2), while the cloud fraction is similar between the simulations (Figs. 9 and 17). In the manuscript we still present the mass flux at 500 mb as it represents the transition between the warm and the cold parts of the clouds.
- 280

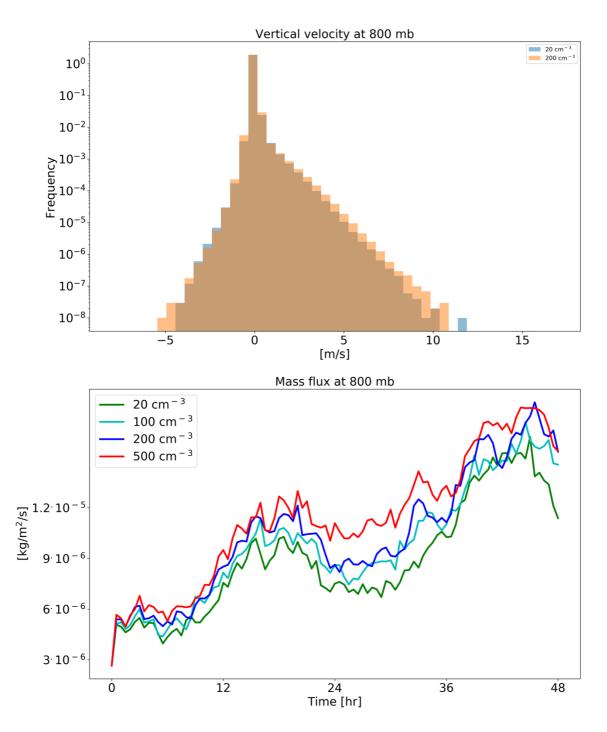


Figure R1. histograms of ICON simulated vertical velocity at the level of 800 mb for a clean (CDNC = 20 cm⁻³) and polluted (CDNC = 200 cm⁻³) simulations (upper), and the time evolution of the net upwards water (liquid and ice) mass flux (lower) for the different CDNC simulations for the shallow-cloud dominated case (10-12/08/2016). In the histogram only two simulations are presented for clarity.

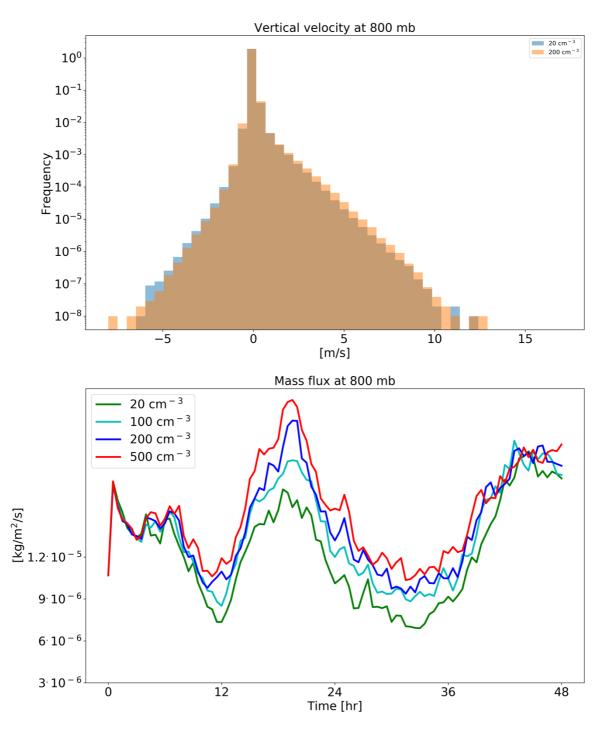


Figure R2. histograms of ICON simulated vertical velocity at the level of 800 mb for a clean (CDNC = 20 cm⁻ 3) and polluted (CDNC = 200 cm⁻³) simulations (upper), and the time evolution of the net upwards water (liquid and ice) mass flux (lower) for the different CDNC simulations for the deep-cloud dominated case (16-18/08/2016). In the histogram only two simulations are presented for clarity.

292

293 (3) Large contribution from residuals

A large contribution to the overall energy budget is within the residual term, which the authorsstate should reduce to zero given a long enough averaging time. How can the authors be sure

that this is true and that the large component in the residual term is not a "buffering" effect -

e.g. changing stability of the atmosphere to compensate for the changing energy budget? Canthe 3D distribution of the residual values be used to quantify this at all?

Reply: A similar argument was raised by reviewer #1 – we will repeat our reply here. What was previously called a "residual" term was changes in the revised manuscript to be refer to as "energy imbalance" as it better describes it. A recent study shows that in order to get close to energy balance the spatial scale should be on the order of ~5000km (Jakob et al., 2019) and the time scale longer than a month (we found a similar scale using GCMs). Our simulations operate on smaller spatial-temporal scales than that and hence it is not surprising that we obtain an imbalance.

The energy imbalance is composed of changes in the storage term and local divergence or convergence of dry-static energy into the domain. In our case, almost the entire imbalance is simply dry static energy that moves in or out of the domain. For example, in the deep-cloud dominated case there is a net production of dry-static energy in the domain by precipitation (which is not entirely balanced by the radiative cooling). This extra dry-static energy is then advected out of the domain.

312 An explanation about this point was added to the revised manuscript:

313 *"The total column atmospheric energy budget can be described as follows:*

 $314 \qquad LP + Q_R + Q_{SH} = div(s) + ds/dt \qquad (1)$

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)."

322

"In this shallow-cloud dominated case the radiative cooling of the atmosphere is significantly
larger than the warming due to precipitation (mean of -114.7 W/m² compared to 90.1 W/m²),
hence the energy imbalance (R) is negative. Negative R means that there must be some
convergence of dry static energy into the domain and/or decrease in the storage term, in this
case it is mostly due to convergence of dry static energy."

- "Next, we analyse the atmospheric energy budget for the deep-cloud dominated case (Fiona tropical storm Fig. 12). As opposed to the shallow-cloud dominated case, in this case the LP contribution dominates over the radiative cooling and hence the energy imbalance R is positive and large, suggesting divergence of dry static energy out of the domain."
- 333
- (4) There appears to be a mismatch between TWP in Figure 8 (lower right plot) and qt in Figure 334 9 (lower central plot). Similarly in Figures 16 & 17. The vertical profile of qt is quite similar 335 for 3 simulations in the shallow case (Figure 9; excluding the 500 cm-3 line). Similarly, the qt 336 values from 3 simulations in the deep case are also similar (Figure 17; excluding the 20 cm-3 337 line). However, in figure 8 & 16 these is clear separation between all the TWP lines throughout 338 the simulation. By quick calculation the spread in the TWP timeseries seems too large to be 339 explained by the differences in qt (which are mostly between 650-400 hPa). How can this 340 341 difference be explained? Is the TWP only including cloud and ice, but ignoring rain water? Similarly, is the LW only cloud, ignoring rain? 342
- Reply: Thank you for this comment that helped us clarify this point. Indeed, the LWP include the cloud mass (qc) and not the rain mass (qr). This is done for consistency with LWP calculated from satellite observations, which are sensitive only to the cloud mass and not to the rain mass (see also the reply to the next point). This is now better explained in the revised manuscript:
- ³⁴⁸ "Figure 8. Domain average properties as a function of time for the different CDNC simulations for the ³⁴⁹ shallow-cloud dominated case. The properties that are presented here are: cloud fraction (CF), rain rate, ³⁵⁰ temperature in 2 m, liquid water path (LWP – based on the cloud water mass, excluding the rain mass for ³⁵¹ consistency with satellite observations), ice water path (IWP) and total water path (TPW = LWP + IWP). For ³⁵² each property, the mean difference between all combinations of simulations, normalized to a factor 5 increase ³⁵³ in CDNC, and its standard deviation appear in parenthesis."
- 354
- (5) Following from the above point: is rain water radiatively interactive in the model? If not,
 to what extent does this removal of mass from the radiatively interactive cloud species have on
 the Twomey effect calculations performed, given that the rain water mass is almost equal to
- 358 the cloud water mass at some heights?
- <u>Reply:</u> As done in most atmospheric models (Hill et al., 2018), the rain mass is not included in
 the radiative calculations. Since the rain drops are much larger than the cloud droplet, their
- 361 cross-section available for interaction with radiation (for a given water mass) is much smaller

and usually negligible (Hill et al., 2018). For example, a simple "back of the envelop" calculation of the cloud optical depth (τ) as in Heus & Seifert (2013) and Spill et al. (2019) follows as:

365 $\tau = 0.19 * LWP^{5/6} * N^{1/3}$,

where LWP is the liquid water path and N is the drop concentration, for a given LWP for cloud 366 droplets with radius r=10 µm and for rain drops with r=0.5 mm yield a factor of 50 decrease in 367 τ for the rain compare with the cloud. Generally, as τ is proportional to N^{1/3} it decreases 368 proportional to r (for a given LWP), and since the cloud droplets and rain drops are separated 369 by 1 or 2 orders of magnitudes, the effect of the cloud droplets on the radiation is much larger 370 than that of the rain drops. This simple calculation does not account for the changes in Mie size 371 parameter between rain drops and cloud droplets but it serves to demonstrate the orders of 372 magnitude differences between the two different regimes. 373

374

375 (6) Impact of simplifications

The approach of simply modifying the CDNC instead of the aerosol concentration of the atmosphere ignores several potentially important processes/feedbacks (e.g. activation of

378 CCN/IN, size distribution of aerosol, direct radiative effects) - the authors should comment on

379 these shortcomings in the conclusions.

380 <u>Reply:</u> Thank you. Based on this comment we have added a comment about the limitation of

using a fixed CDNC simulations:

- 382 "The different CDNC scenarios serve as a proxy for different aerosol conditions (as the first order effect of increased aerosol concentration on clouds is to increase the CDNC, Andreae, 383 384 2009). This also allows to separate the cloud response from the uncertainties involved in the representation of the aerosols in numerical models (Ghan et al., 2011; Simpson et al., 2014; 385 386 Rothenberg et al., 2018). However, it limits potential feedbacks between clouds and aerosols, such as the removal of aerosol levels by precipitation scavenging and potential aerosol effects 387 thereon. In addition, the fixed CDNC framework does not capture the differences in aerosol 388 activation between shallow and deep clouds, due to differences in vertical velocity. Another 389 aerosol effect that is not included in our simulations is the direct interaction between aerosol 390 and radiation. In future work we plan to examine the mutual interaction between microphysical 391 effects and the direct aerosol radiative effects." 392
- 393
- 394

395 In the conclusions:

396 *"Furthermore, we do not include the temporal evolution of the aerosol concentration.*

397 *Feedbacks between the aerosol concentration and clouds processes (such as wet scavenging),*

- as well as the direct effects of aerosol on radiation would add another layer of complexity that
- *should be accounted for in future work.*"
- 400

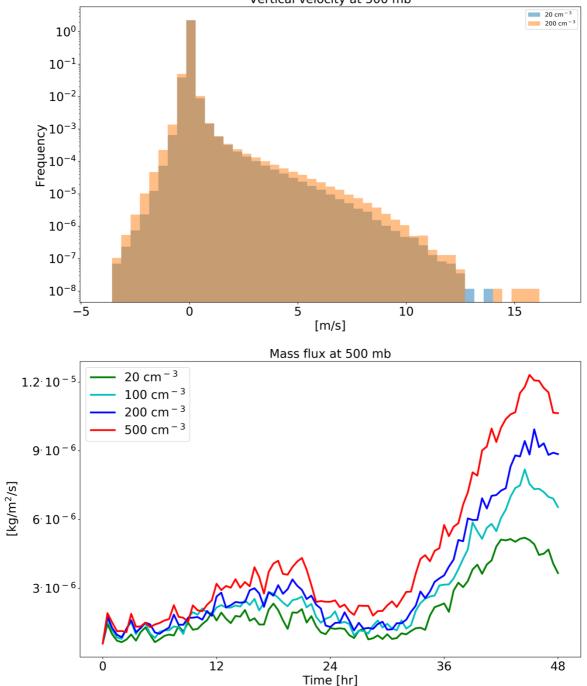
401 (7) Robustness of results

The authors should comment on the robustness of these results, in light of the fact that single simulations (rather than ensembles) of two individual case studies are performed. The results in figures 8 & 16 suggest a clear separation between all 4 CDNC concentrations from early on in the simulation - however, the vertical profiles of qi, qt and CF in figures 9 & 17 suggest that the 20 CDNC cm-3 simulation is the only one of the four that is substantially different (particularly at upper levels, which seem to be most important in this story).

Reply: The robustness of our simulations, which, as the reviewer stated, are based on few 408 simulations rather then on large ensemble, occupied our mind as well. In a recent paper which 409 is currently under discussion in ACPD (Dagan and Stier, 2019) we investigate this exact 410 question. In that paper we use a smaller domain compared to the current study (3° x 3° rather 411 412 than 22° x 11°) to simulated a large ensemble of initial conditions (all together we simulate 124 different simulations). This large ensemble enables robust identification of the effect of CDNC 413 414 changes on cloud properties. We were able to show that the general conclusions which are stated in the current study hold also for large statistics. Based on the following comment we 415 416 have added a discussion about the robustness of our results to the revised manuscript:

"There exists a large spread in estimates of aerosol effects on clouds for different cloud types 417 and different environmental conditions. In this study, as we use a relatively large domain (22° 418 x 11°) and two different dates (each for two days), we sample many different local 419 420 environmental conditions and cloud types. Such more realistic setups (although with lower spatial resolution) could provide more reliable estimates of aerosol effects on heterogeneous 421 cloud systems than just one-cloud-type, small domain simulations (as was done in many 422 previous studies, e.g (Dagan et al., 2017; Seifert et al., 2015; Ovchinnikov et al., 2014)). 423 However, the conclusions demonstrated here are based on two specific cases. In order to 424 examine the validity of our main conclusions over a wider range of initial conditions, we have 425 conducted a large ensemble of simulations starting from realistic initial conditions (although 426 with a smaller domain) in a companion paper (Dagan and Stier, 2019). These simulations 427

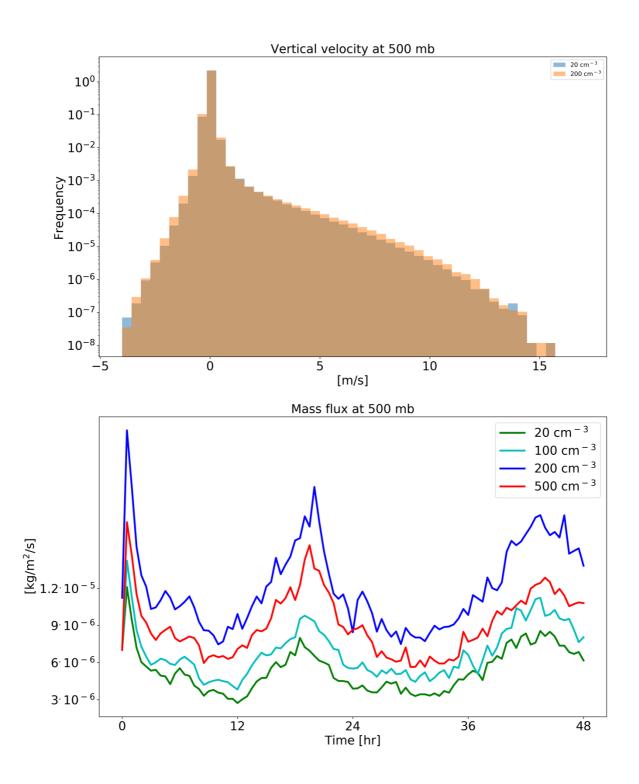
- 428 *demonstrate that the main conclusions presented in this paper are robust and hold also for a*
- 429 wide range of initial conditions representative for this area."
- 430
- 431 Minor points: please show the mass flux from all four simulations in figures 11 and 19 to be
- 432 consistent with the other plots in the paper.
- 433 <u>Reply:</u> The mass flux of all four simulations are now presented in Figs. 11 and 19:



Vertical velocity at 500 mb

Figure 11. histograms of ICON simulated vertical velocity at the level of 500 mb for a clean (CDNC = 20 cm⁻³) and polluted (CDNC = 200 cm⁻³) 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 (10-12/08/2016). The 500 mb level is chosen as it represents the transition between the warm part to the cold part of the clouds. In the histogram only two simulations are presented for clarity.

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- 441



443 Figure 19. histograms of ICON simulated vertical velocity at the level of 500 mb for a clean (CDNC = 20 cm⁻³) 444 and polluted (CDNC = 200 cm⁻³) simulations (upper), and the time evolution of the net upwards water (liquid 445 and ice) mass flux (lower) for the different CDNC simulations for the deep-cloud dominated case (16-446 18/08/2016). The 500 mb level is chosen as it represents the transition between the warm part to the cold part 447 of the clouds. In the histogram only two simulations are presented for clarity.

448

449

Lines 335-338: please be more quantitative about the results of the test with the offline radiation 450

calculations as to the relative contributions of the cloud fraction and TWP changes. Line 367: 451

please quantify the "vast majority" of LW flux changes due to cloudy rather than clear skies. 452

Reply: Thank you. More information was added to the revised manuscript: 453

"For estimating the relative contribution of the changes in CF and water content to the SW 454 flux changes we have conducted off-line radiative transfer sensitivity tests. To quantify the 455 water content radiative effect, we feed the same CF vertical profile from the model into the 456 offline radiative transfer model BUGSrad, while allowing the water content vertical profile to 457 change (and visa versa to compute the CF radiative effect). This approach demonstrates that 458 the contribution from the small reduction in CF is negligible compared to the increased SW 459 reflectance caused by the increased water content (the effect of the reduction in CF compensate 460 only about 1% of the effect of the increase in the water content)." 461

462

"The increased humidity at the upper troposphere would act to decrease the outgoing LW flux, 463 similar to the effect of the increased ice content in the upper troposphere (Fig. 9). However, 464 sensitivity studies with off-line radiative transfer calculations using BUGSrad demonstrate that 465 the vast majority (more than 99%) of the different in F_{LW}^{TOA} between clean and polluted 466 conditions emerges from the cloudy skies (rather than clear-sky), suggesting that the effect of 467 the increased ice content at the upper troposphere dominates." 468

469

The plots in figures 10 and 18, currently described in the caption as Hovmöller plots, would be 470 better described as time-height plots.

471

Reply: Thanks. It was changed according to the reviewer's suggestion. 472

473

Is there an explanation for the relative minimum of cloud water content at 650 hPa in all 474

simulations? I struggle to find a physical explanation for this. 475

476 <u>Reply:</u> We think that the relative minimum of domain-wide cloud water at 650 hPa is simply

477 due to the fact that below this level there are still quite a lot of shallow clouds while above it

there are anvils clouds with longer lifetime (and hence the larger cloud water mass) then the

479 liquid at lower levels.

480

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508 Atmospheric energy budget response to idealized aerosol perturbation in

509 **tropical cloud systems**

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517

518 Abstract

519 The atmospheric energy budget is analysed in numerical simulations of tropical cloud systems-This is done in order to better understand the physical processes behind aerosol effects on the 520 atmospheric energy budget. The simulations include both shallow convective clouds and deep 521 convective tropical clouds over the Atlantic Ocean. Two different sets of simulations, at different 522 dates (10-12/8/2016 and 16-18/8/2016), are being simulated with different dominant cloud 523 524 modes (shallow or deep). For each case, the cloud droplet number concentrations (CDNC) is varied as a proxy for changes in aerosol concentrations. It is shown that the total column 525 526 atmospheric radiative cooling is substantially reduced with CDNC in the deep-cloud dominated case (by $\sim 10.0 \text{ W/m}^2$), while a much smaller reduction ($\sim 1.6 \text{ W/m}^2$) is shown in the shallow-527 cloud dominated case. This trend is caused by an increase in the ice and water vapor content at 528 the upper troposphere that leads to a reduced outgoing longwave radiation, an effect which is 529 530 stronger under deep-cloud dominated conditions. A decrease in sensible heat flux (driven by increase in the near surface air temperature) reduces the warming by ~ 1.4 W/m² in both cases. It 531 532 is also shown that the cloud fraction response behaves in opposite ways to an increase in CDNC, showing an increase in the deep-cloud dominated case and a decrease in the shallow-cloud 533 dominated case. This demonstrates that under different environmental conditions the response to 534 aerosol perturbation could be different. 535

536

537 **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/m² (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.

Beside its effect on the radiation budget, aerosols may affect the precipitation distribution and 544 total amount (Levin and Cotton, 2009; Albrecht, 1989; Tao et al., 2012). A useful perspective to 545 improve our understanding of aerosol effect on precipitation, which became common in the last 546 few years, arises from constraints on the energy budget (O'Gorman et al., 2012; Muller and 547 O'Gorman, 2011; Hodnebrog et al., 2016; Samset et al., 2016; Myhre et al., 2017; Liu et al., 548 2018; Richardson et al., 2018; Dagan et al., 2019a). On long time scales, any precipitation 549 perturbations by aerosol effects will have to be balanced by changes in radiation fluxes, sensible 550 heat flux or by divergence of dry static energy. The energy budget constraint perspective was 551 552 found useful to explain both global (e.g. (Richardson et al., 2018)) and regional (Liu et al., 2018; Dagan et al., 2019a) precipitation response to aerosol perturbations in global scale simulations. 553 In this study, we investigate the energy budget response to aerosol perturbation on a regional 554 scale using high resolution cloud resolving simulations. This enables an improved understanding 555 of the microphysical processes controlling atmospheric energy budget perturbations. The strong 556 connection between the atmospheric energy budget and convection has long been appreciated 557 (e.g. (Arakawa and Schubert, 1974; Manabe and Strickler, 1964)) as well as the connection to 558 the general circulation of the atmosphere (Emanuel et al., 1994). 559

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

561
$$LP + Q_R + Q_{SH} = \operatorname{div}(s) + \operatorname{ds/dt}$$
 (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 <u>the</u> 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 <u>energy imbalance (which is</u> <u>calculated as the residual [(R]) of the left-hand side)</u>.

569 Q_R is defined as:

570 $Q_R = (F_{SW}^{TOA} - F_{SW}^{SFC}) + (F_{LW}^{TOA} - F_{LW}^{SFC})$ (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

573 (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 radiationand 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 576 (CCN) and ice nuclei (IN). Larger aerosol concentrations, e.g. by anthropogenic emissions, could 577 lead to larger cloud droplet and ice particle concentrations (Andreae et al., 2004; Twomey, 1977; 578 Hoose and Möhler, 2012). Changes in hydrometer concentration and size distribution were 579 shown to affect clouds' microphysical processes rates (such as condensation, evaporation, 580 581 freezing and collision-coalescence), which in turn could affect the dynamics of the clouds (Khain et al., 2005; Koren et al., 2005; Heikenfeld et al., 2019; Chen et al., 2017; Altaratz et al., 2014; 582 Seifert and Beheng, 2006a), the rain production (Levin and Cotton, 2009; Albrecht, 1989; Tao 583 et al., 2012) and the clouds' radiative effect (Koren et al., 2010; Storelvmo et al., 2011; Twomey, 584 1977; Albrecht, 1989). The aerosol effect, and in particular its effects on the radiation budget 585 586 and the atmospheric energy budget, is cloud regime dependent (Altaratz et al., 2014; Lee et al., 2009; Mülmenstädt and Feingold, 2018; van den Heever et al., 2011; Rosenfeld et al., 2013; 587 Glassmeier and Lohmann, 2016; Gryspeerdt and Stier, 2012; Christensen et al., 2016), time 588 dependent (Dagan et al., 2017; Gryspeerdt et al., 2015; Seifert et al., 2015; Lee et al., 2012; 589 Dagan et al., 2018c), aerosol type and size distribution dependent (Jiang et al., 2018; Lohmann 590 and Hoose, 2009) and (even for a given cloud regime) meteorological conditions dependent 591 (Dagan et al., 2015a; Fan et al., 2009; Fan et al., 2007; Kalina et al., 2014; Khain et al., 2008) 592 and was shown to be non-monotonic (Dagan et al., 2015b; Jeon et al., 2018; Gryspeerdt et al., 593 2019; Liu et al., 2019). Hence the quantification of the global mean radiative effect is extremely 594 challenging (e.g. (Stevens and Feingold, 2009; Bellouin et al., 2019)). 595

Previous studies demonstrated that the mean aerosol effect on deep convective clouds can 596 increase the upward motion of water, and hence also increase the cloud anvil mass and extent 597 (Fan et al., 2010; Chen et al., 2017; Fan et al., 2013; Grabowski and Morrison, 2016). The 598 increase in mass flux to upper levels was explained by the convective invigoration hypothesis 599 (Fan et al., 2013; Koren et al., 2005; Rosenfeld et al., 2008; Seifert and Beheng, 2006a; Yuan 600 et al., 2011a; Williams et al., 2002), which was proposed to lead to stronger latent heat release 601 under higher aerosol concentrations and hence stronger vertical velocities. In addition to the 602 stronger vertical velocities, under polluted conditions the smaller hydrometers are being 603 transported higher in the atmosphere (for a given vertical velocity (Chen et al., 2017; Koren et 604 al., 2015; Dagan et al., 2018a)) and their lifetime at the upper troposphere is longer (Fan et al., 605 606 2013; Grabowski and Morrison, 2016). The invigoration mechanism can also lead to an increase

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
by latent heat release, and the increased anvil mass and extent by longwave radiative warming
(Koren et al., 2010; Storelvmo et al., 2011). However, it should be pointed out that the
uncertainty underlying these proposed effects remain significant (White et al., 2017; Varble,
2018). In addition, aerosol effects on precipitation from deep convective cloud was shown to be
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 614 (Dagan et al., 2015a; Dagan et al., 2017). However, unlike in the deep clouds case, the mean 615 effect on precipitation, under typical modern-day conditions, is thought to be negative (Albrecht, 616 1989; Rosenfeld, 2000; Jiang et al., 2006; Xue and Feingold, 2006; Dagan and Chemke, 2016). 617 The aerosol effect on shallow cloud cover and mean water mass (measure by liquid water path -618 619 LWP) might also depend on the meteorological conditions and aerosol range (Dagan et al., 2015b; Dagan et al., 2017; Gryspeerdt et al., 2019; Dey et al., 2011; Savane et al., 2015) and is 620 621 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 622 623 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 624 cloud coverage (Small et al., 2009; Jiang et al., 2006; Costantino and Bréon, 2013; Seigel, 625 2014)). Another addition to this complex response is the fact that the aerosol effect on warm 626 convective clouds was shown to be time dependent and affected by the clouds' feedbacks on the 627 thermodynamic conditions (Seifert et al., 2015; Dagan et al., 2016; Dagan et al., 2017; Lee et al., 628 2012; Stevens and Feingold, 2009; Dagan et al., 2018b). Previous simulations that contained 629 several tropical cloud modes demonstrate that increase in aerosol concentrations can lead to 630 suppression of the shallow mode and invigoration of the deep mode (van den Heever et al., 2011). 631 Hence the domain mean effect, even if it is demonstrated to be small, may be the result of 632 opposing relatively large contributions from the different cloud modes (van den Heever et al., 633 634 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 635 636 precipitation changes.

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

640 perspective in constraining aerosol effect on precipitation. However, the physical processes 641 behind aerosol-cloud microphysical effects on the energy budget are still far from being fully 642 understood. In this study we use cloud resolving simulations to increase our understanding of the 643 effect of microphysical aerosol-cloud interactions on the atmospheric energy budget.

644 Methodology

645 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 646 647 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 648 649 et al., 2014; Stevens et al., 2019; Stevens et al., 2016)) campaign, which took place during August 2016 in the western part of the northern tropical Atlantic. We use existing NARVAL 2 650 convection-permitting simulations (Klocke et al., 2017) as initial and boundary conditions for 651 our simulations. 652

The domain covers ~22° in the zonal direction (25° - 47° W) and ~11° in the meridional direction 653 (6° - 17° N) and therefore a large fraction of the northern tropical Atlantic (Fig. 1). During August 654 2016, the intertropical convergence zone (ITCZ) was located in the southern part of the domain 655 while the northern part mostly contains trade cumulus clouds. Hence, this case study provides 656 an opportunity to study heterogenous clouds systems. Daily variations in the deep/shallow cloud 657 658 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 659 mode (10-12/8/2016 - see Fig. 2, and Figs S1 and S3, supporting information-SI), and one that 660 represents a deep-cloud dominated mode (16-18/8/16 - see Fig. 3 and Figs. S2 and S3, SI). In 661 the shallow-cloud dominated case, most of the domain is covered by trade cumulus clouds that 662 are being advected with the trade winds from north-east to south-west. In the southern part of the 663 domain, throughout most of the simulation, there is a zonal band of deep convective clouds (Fig. 664 2) that contribute on average ~25% out of the total cloud cover (Fig. S3, SI). The deep-cloud 665 dominated case represents the early stages of the development of the tropical storm Fiona (Fig. 666 3). Fiona formed in the eastern tropical Atlantic and moved toward the west-north-west. It started 667 as a tropical depression at 16/8/2016 18:00 UTC while its centre was located at 12.0° N 32.2° W. 668 It kept moving towards the north-west and reach a level of a tropical storm at 17/8/2016 12UTC, 669 while located 13.7° Ν 670 its centre was at 36.0° W (https://www.nhc.noaa.gov/data/tcr/AL062016 Fiona.pdf). The general propagation speed and 671

direction, strength (measure by maximal surface wind speed) and location of the storm are predicted well by the model. However, the model produces more anvil clouds than what was observed from the satellite (Fig. 3). These two different cases, representing different atmospheric energy budget initial state (see also Figs. 4 and 12 below), enable the investigation of the aerosol effect on the energy budget under different initial conditions.

We use a two-moment bulk microphysical scheme (Seifert and Beheng, 2006b). For each case, 677 four different simulations with different prescribed cloud droplet number concentrations 678 (CDNC) of 20, 100, 200, and 500 cm⁻³ are conducted. The different CDNC scenarios serve as 679 a proxy for different aerosol-concentration conditions (as the first order effect of increased 680 aerosol concentration on clouds is to increase the CDNC, (Andreae, 2009)). This also allows 681 682 to separate the cloud response from and avoid the uncertainties involved in the representation of the aerosols in numerical models (Ghan et al., 2011; Simpson et al., 2014; Rothenberg et al., 683 2018). However, it limits potential feedbacks between clouds and aerosols, such as the removal 684 685 of aerosol levels by precipitation scavenging and potential aerosol effects thereon. In addition, the fixed CDNC framework does not capture the differences in aerosol activation fraction 686 between shallow and deep clouds, due to differences in vertical velocity. Another aerosol effect 687 that is not included in our simulations is the direct interaction between aerosol and radiation. 688 In future work we plan to examine the mutual interaction between the microphysical effects 689 690 and the direct aerosol radiative effects.

For calculation of the difference between high CDNC (polluted) conditions and low CDNC 691 (clean) conditions, the simulations with CDNC of 200 and 20 cm⁻³ are chosen as they represent 692 the range typically observed over the ocean (see for example the CDNC range presented in 693 694 recent observational-based studies (Rosenfeld et al., 2019; Gryspeerdt et al., 2019)). Each simulation is conducted for 48 hours starting from 12 UTC. The horizontal resolution is set to 695 1200 m and 75 vertical levels are used. The temporal resolution is 12 sec and the output interval 696 is 30 min. Interactive radiation is calculated every 12 min using the RRTM-G scheme (Clough 697 et al., 2005; Iacono et al., 2008; Mlawer et al., 1997). We have added a coupling between the 698 microphysics and the radiation to include the Twomey effect (Twomey, 1977). This was done 699 by including the information of the cloud liquid droplet effective radius, calculated in the 700 microphysical scheme, in the radiation calculations. No Twomey effect due to changes in the 701 ice particles size distribution was considered due to the large uncertainty involved in the ice 702 microphysics and morphology. Additional details, such as the surface and atmospheric physics 703

parameterizations, are described in Klocke et al., (2017) and include an interactive surface flux
scheme and a fixed sea surface temperature (SST). We note that using a fixed SST does not
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
change over the two days simulations.

For comparing the outgoing longwave flux from the simulations and observations we use 709 imager data from the SEVIRI instrument onboard the Meteosat Second Generation (MSG) 710 geostationary satellite (Aminou, 2002). The outgoing longwave flux is calculated using the 711 Optimal Retrieval for Aerosol and Cloud (ORAC) algorithm (Sus et al. 2017; McGarragh, et 712 al. 2017). Cloud optical (thickness, effective radius, water path) and thermal (cloud top 713 714 temperature and pressure) properties are retrieved from ORAC using an optimal estimationbased approach. These retrievals and reanalysis profiles of temperature, humidity and ozone 715 are then ingested into BUGSrad, a two-stream correlated-k broadband flux algorithm (Stephens 716 et al., 2001) that outputs the fluxes at the top and bottom of the atmosphere and shown to have 717 excellent agreement when applied to both active (CloudSat) and passive (Advanced Along 718 Track Scanning Radiometer) satellite sensors compared to Clouds and the Earth's Radiant 719 Energy System (Henderson et al. 2013; Stengel et al. 2019). In addition, off-line sensitivity 720 radiative transfer tests using vertical profiles from our model were conducted with BUGSrad 721 to identify the source of the differences in fluxes between clean and polluted conditions. 722

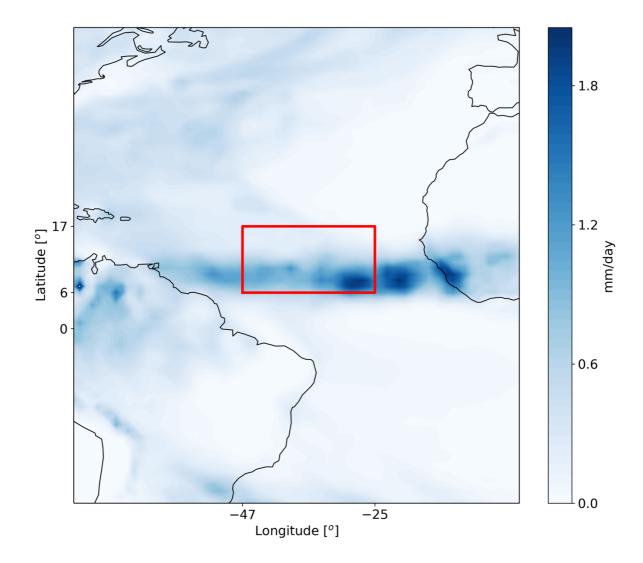
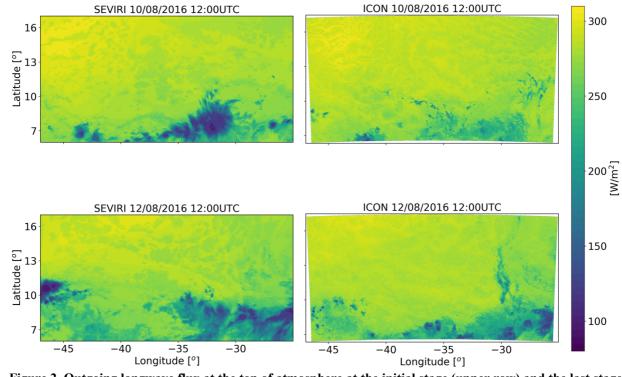
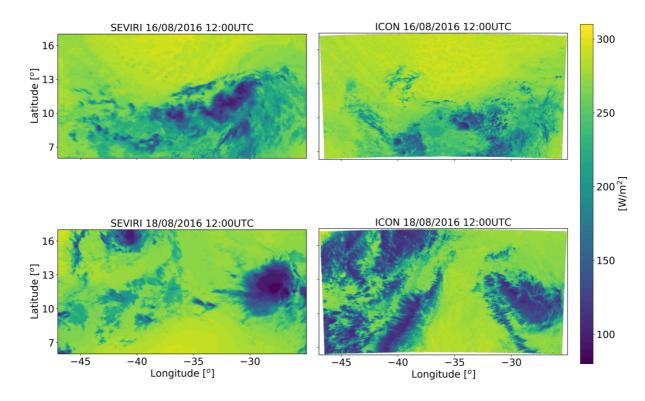


Figure 1. Domain of the ICON simulations (red rectangle) for the NARVAL 2 case study overlaid on the

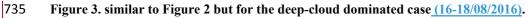
August 2016 ECMWF era-interim reanalysis (Dee et al., 2011) mean precipitation rate.



Longitude [°]
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 (1012/08/2016) from geo-stationary satellite (SEVIRI-MSG – right column) and the ICON model simulation with
CDNC of 20 cm⁻³ (left column).



734



736 **Results**

737 Shallow-cloud dominated case -10-12/08/2016

We start with energy budget analysis of the shallow-cloud dominated case base simulations 738 $(CDNC = 20 \text{ cm}^{-3})$. Figure 4 presents the time mean (over the two days simulation) of the 739 different terms of the energy budget (Equation 1). As expected, LP dominates the warming of 740 the atmosphere while Q_R dominate the cooling. The sensible heat flux (Q_{SH}) is positive (act to 741 warm the atmosphere) but it is an order of magnitude smaller than the LP and Q_R magnitudes. In 742 this shallow-cloud dominated case the radiative cooling of the atmosphere is significantly larger 743 than the warming due to precipitation (mean of -114.7 W/m² compared towith 90.1 W/m²), hence 744 the energy imbalance residual (R) is negative. Negative R means that there must be some 745 746 convergence of dry static energy into the domain and/or decrease in the storage term, in this case it is mostly due to convergence of dry static energy. 747

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

- 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 W/m²
- 752 (1 mm/hr of precipitation is equivalent to 628 W/m^2), however, the vast majority of the domain

contributes very little in terms of *LP*. Q_R 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.

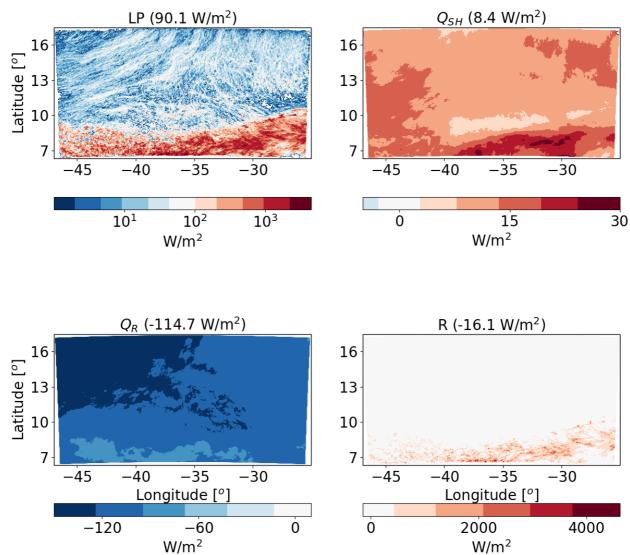
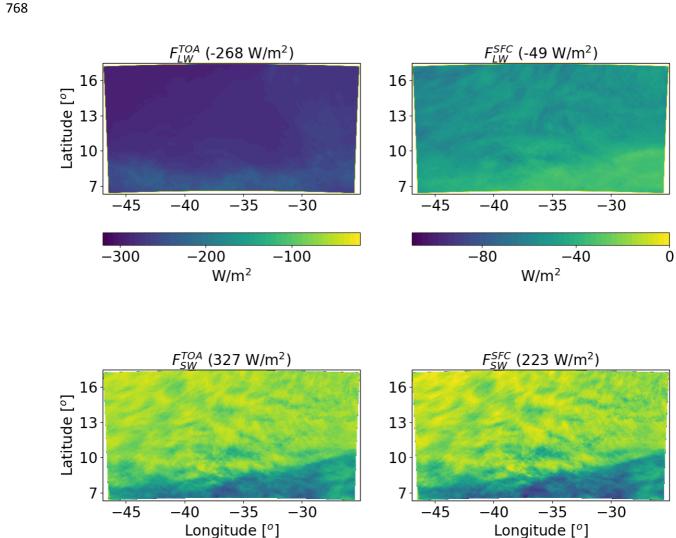


Figure 4. Spatial distribution of the time mean of the different terms of the energy budget for the ICON simulation of the shallow-cloud dominated case (10-12/08/2016) with CDNC = 20 cm⁻³. The terms that appear here are: *LP* - latent heat by precipitation, Q_{SH} - sensible heat flux, Q_R - atmospheric radiative warming, and R – the residual 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 lower 766 mean cloud fraction) in the rest of the domain. 767



W/m² 769 770 Figure 5. Spatial distribution of ICON simulated time-mean longwave (LW) and shortwave (SW) radiation 771 fluxes at the top of atmosphere (TOA) and surface (SFC) for a simulation of the shallow-cloud dominated 772 case (10-12/08/2016) with CDNC = 20 cm⁻³. The domain and time mean value of each term appears in 773 parenthesis.

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Response to aerosol perturbation – shallow-cloud dominated case 775

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W/m²

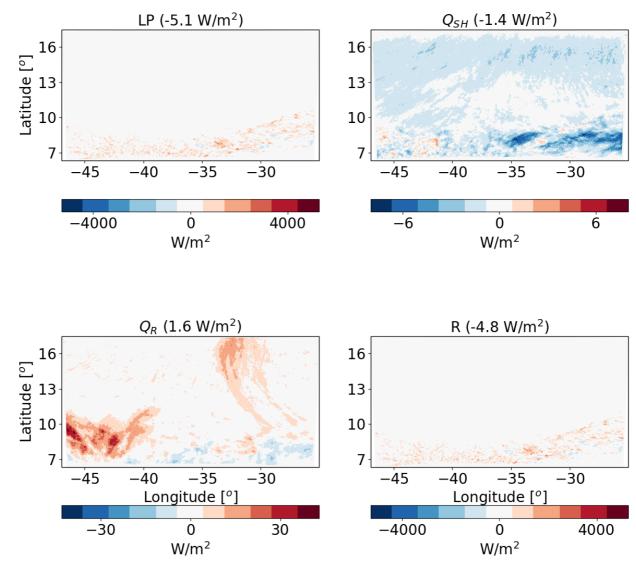
Next, we analyse the response of the atmospheric energy budget of this case to perturbations in 776 777 CDNC. Figure 6 presents the differences in the different terms of the energy budget between a polluted simulation (CDNC = 200 cm^{-3}) and a clean simulation (CDNC = 20 cm^{-3}). It 778

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200

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demonstrates that the LP differences between the different CDNC scenarios contribute 5.1 W/m² 779 less to warm the atmosphere in the polluted vs. the clean simulation. We note that this apparently 780 large effect is caused by a small, non-statistically significant, precipitation difference (~0.4 mm 781 over the two days of simulation - see Fig. 8 below). The strong sensitivity of the atmospheric 782 energy budget to small precipitation changes (recalling that 1 mm/hr is equivalent to 628 W/m²) 783 exemplifies the caution one needs to take when looking on precipitation response in terms of 784 energy budget perspective. The Q_R differences lead to relative warming of the atmosphere of the 785 polluted case compared to the clean case by 1.6 W/m². We note that most of the Q_R differences 786 are located in the south-west part of the domain. The Q_{SH} changes counteracts 1.4 W/m² of the 787 atmospheric warming by Q_R and so the end result is a deficit of 4.8 W/m² in the atmospheric 788 energy budget in the polluted simulation compared to the clean simulation. The decrease in the 789 Q_{SH} is driven by an increase in the near surface air temperature (see Fig. 8). 790

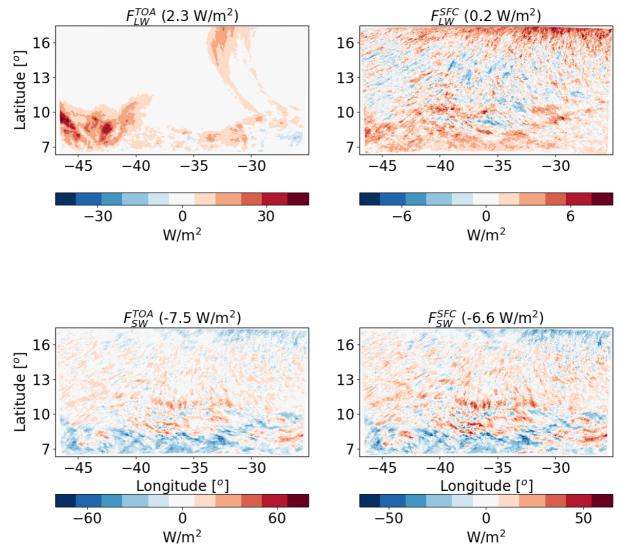


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Figure 6. The differences between polluted (CDNC = 200 cm⁻³) and clean (CDNC = 20 cm⁻³) ICON simulations of the time-mean terms of the energy budget for the shallow-cloud dominated case (10-12/08/2016). The terms that appears here are: *LP* - latent heat by precipitation, Q_{SH} - sensible heat flux, Q_R - atmospheric radiative warming, and R – the residual 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 798 the different radiative fluxes. Figure 7 demonstrates that most of the relative atmospheric 799 radiative heating in the polluted case compared to the clean case is contributed by changes in the 800 F_{LW}^{TOA} fluxes. The changes in F_{LW}^{SFC} are an order of magnitude smaller. The SW fluxes change both 801 at the TOA and SFC are larger than the F_{LW}^{TOA} changes, however, in terms of the atmospheric energy 802 budget, they almost cancel each other out and the net SW atmospheric effect is only -0.9 W/m². 803 Most of the reduction in SW fluxes (both at TOA and the surface) comes from the deep 804 convective regions in the south of the domain while the shallow cloud regions experience some 805

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/m².



W/m² W/m² Figure 7. The differences between polluted (CDNC = 200 cm⁻³) and clean (CDNC = 20 cm⁻³) 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.

815

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⁻³ (on top of the two that were examine above -20 and 820 200 cm⁻³). Figure 8 demonstrates that the domain mean cloud fraction (CF) generally decreases 821 with the increase in CDNC (except for the first ~10 hours of the simulations). Examining the 822 vertical structure of the CF response (Fig. 9), demonstrates that with the increase in CDNC there 823 is a reduction in the low level (below 800 mb) CF concomitantly with an increase in CF at the 824 825 middle and upper troposphere. The differences in rain rate between the different simulations are small. However, both the liquid water path (LWP) and the ice water path (IWP) show a consistent 826 increase with CDNC. Accordingly, also the total water path (TWP), which is the sum of the LWP 827 828 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 829 smaller than 40 µm) increases with CDNC, while the rain mass mixing ratio (qr - drops with 830 radius larger than 40 µm) decreases due to the shift in the droplet size distribution to smaller 831 sizes under larger CDNC conditions. As this case is dominated by shallow clouds, there exists 832 only a comparably small amount of ice mixing ration (qi) (c.f. Fig. 17), but its concentration 833 increases with the CDNC increase. The combined effect of the increase in CDNC is to 834 monotonically increase the total water mixing ratio (qt) above 800 mb (Fig. 9). The relative 835 increase in qt with CDNC becomes larger at higher levels. 836

837 The increase in cloud water with increasing CDNC can explain both the reductions in the net downward SW fluxes (both at TOA and surface) and the decrease in outgoing LW flux at TOA 838 839 (Fig. 7), as it results in more SW reflection concomitantly with more LW trapping in the 840 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 841 842 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 843 844 relative contribution of the Twomey effect compare to the cloud adjustments (CF and TWP effects) to the SW flux changes, we have re-run the simulations with the Twomey effect turned 845 846 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 847 848 is only -1.7 W/m² as compared to -7.5 W/m² with the Twomey effect, demonstrating the predominant role of the Twomey effect. For estimating the relative contribution of the changes 849 in CF and TWP-water content to the SW flux changes we have conducted off-line radiative 850 transfer sensitivity tests. To quantify the water content TWP radiative effect, we feed the same 851 CF vertical profile from the model into the offline radiative transfer model BUGSrad, while 852

allowing the <u>water content</u>TWP vertical profile to change (and visa versa to compute the CF radiative effect). This approach demonstrates that the contribution from the small reduction in CF is negligible compared to the increased SW reflectance caused by the increase<u>d</u>-in <u>water</u> <u>contentTWP (the effect of the reduction in CF compensate only about 1% of the effect of the</u> <u>increase in the water content</u>).

We also note a monotonic increase in the near surface temperature with CDNC (see also Fig. 10 858 below). This trend can be explained by warm rain suppression with increasing CDNC that leads 859 860 to less evaporative cooling (see the decrease in the total amount of water mass mixing ration just above the surface in Fig. 9, (Dagan et al., 2016; Albrecht, 1993; Seigel, 2014; Seifert and Heus, 861 2013; Lebo and Morrison, 2014)). In addition, it was shown that under polluted conditions the 862 rain drops below cloud base are larger, hence evaporating less efficiently (Lebo and Morrison, 863 2014; Dagan et al., 2016). The increase in the near surface temperature drives the decrease in the 864 865 Q_{SH} (Fig. 6).

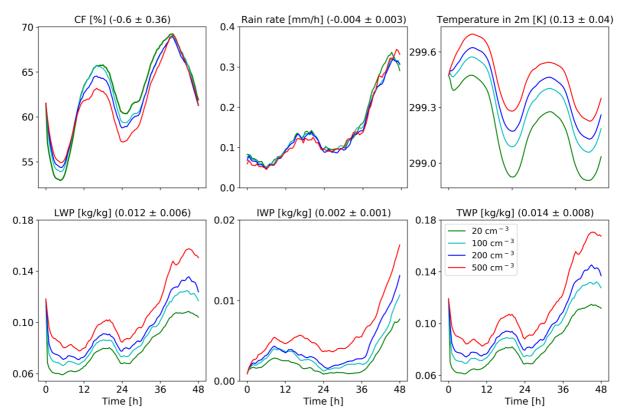
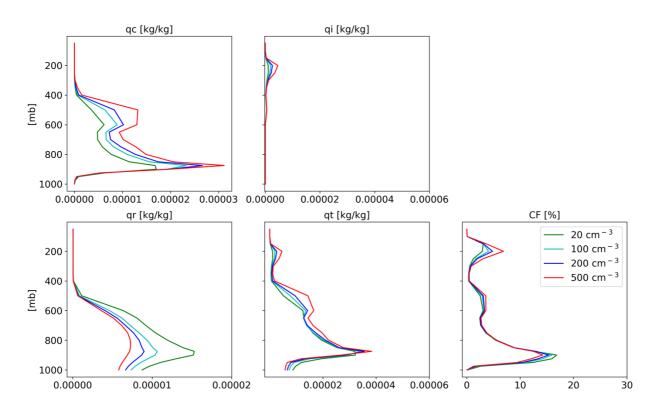




Figure 8. Domain average properties as a function of time for the different CDNC simulations for the shallowcloud dominated case. The properties that are presented here are: cloud fraction (CF), rain rate, temperature in 2 m, liquid water path (LWP <u>– based on the cloud water mass, excluding the rain mass for consistency</u> with satellite observations), ice water path (IWP) and total water path (TPW = LWP + IWP). For each property₂ the mean difference between all combinations of simulations, normalized to a factor 5 increase in CDNC, and its standard deviation appear in parenthesis.

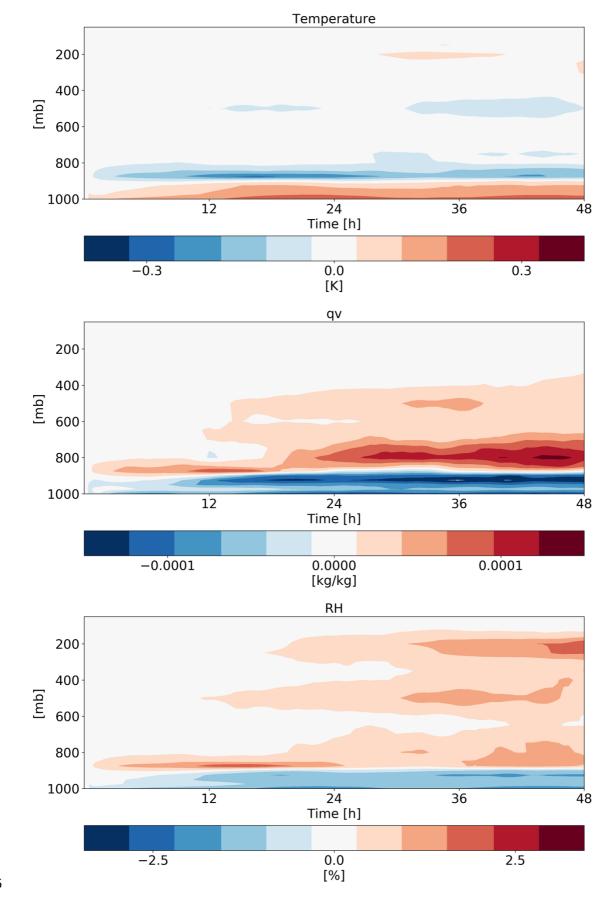


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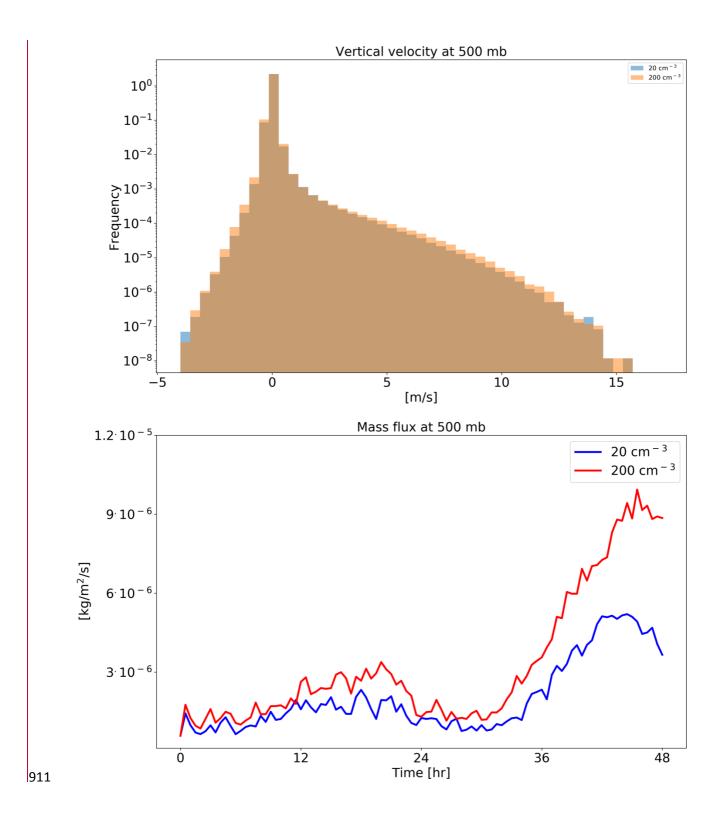
Figure 9. Domain and time average vertical profiles for the different CDNC simulations for the shallow-cloud
dominated case. The properties that are presented here are: cloud droplet mass mixing ratio (qc – for clouds'
droplets with radius smaller than 40 µm), ice mass mixing ratio (qi), rain mass mixing 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.

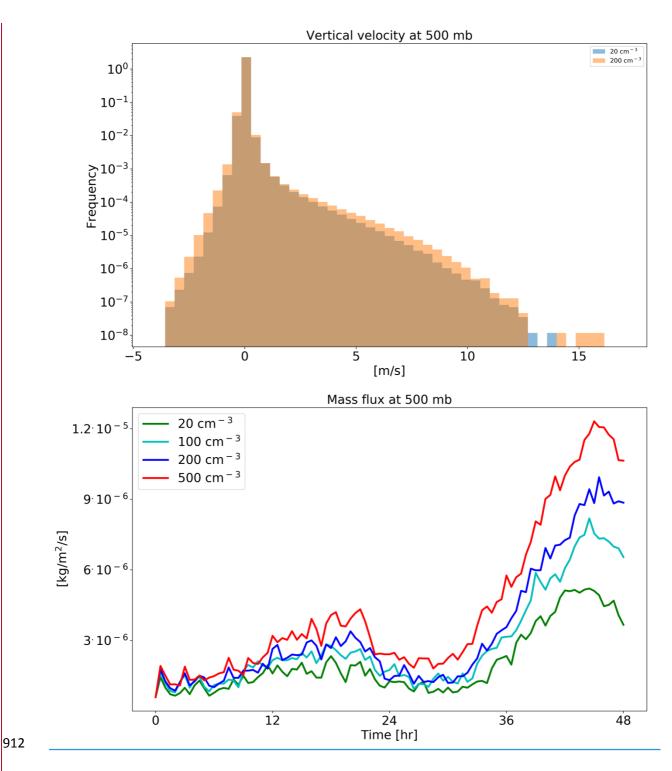
In addition to the clouds' effect on the radiation fluxes, changes in humidity could also contribute 881 (Fig. 10). We note that increase in CDNC leads to increase in relative humidity (RH) and specific 882 humidity (qv) at the middle and upper troposphere without a significant temperature change. The 883 884 increased humidity at the upper troposphere would act to decrease the outgoing LW flux, a similar to the effect ofas the increased ice content inat the upper troposphere-has (Fig. 9). 885 However, sensitivity studies with off-line radiative transfer calculations using BUGSrad, 886 demonstrate that the vast majority (more than 99%) of the different in F_{LW}^{TOA} between clean and 887 polluted conditions emerges from the cloudy skies (rather than clear-sky), suggesting that the 888 effect of the increased ice content at the upper troposphere dominatespredominant. 889

Both the increase in water vapor and ice content <u>inat</u> the upper troposphere are driven by <u>an</u> increase in <u>upward</u> water (liquid and ice) mass flux with increasing CDNC to these levels (Fig. 892 11). An increase 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, 893 tThe increase in mass flux is driven partially by the small increase in vertical velocity 894 (especially for updraft between 5 and 10 m/s - Fig. 11), partially by the small increase in cloud 895 faction at this level (Fig. 9) and mostly due to the larger water mass mixing ratio (Fig. 9) that 896 leads to an increase in mass flux even for a given vertical velocity. The increased relative 897 898 humidity at the upper troposphere, further increases the ice particle lifetime at these levels (in addition to the microphysical effect (Grabowski and Morrison, 2016)) as the evaporation rate 899 900 decreases. In addition, the differences in the thermodynamics evolution between the different simulations (Fig. 10) demonstrate drying and warming of the boundary layer with increasing 901 CDNC, due to reduction in rain evaporation below cloud base and deepening of the boundary 902 layer (Dagan et al., 2016; Lebo and Morrison, 2014; Seifert et al., 2015; Spill et al., 2019). The 903 drying of the boundary layer could explain the reduction in the low cloud fraction (Fig. 9 (Seifert 904 et al., 2015)). 905



- 907 Figure-_10. <u>Time-heightHoymöller</u> diagrams of the differences in the domain mean temperature, specific
- 908 humidity (qv) and relative humidity (RH) vertical profiles between polluted (CDNC = 200 cm⁻³) and clean
- 909 (CDNC = 20 cm^{-3}) simulations for the shallow-cloud dominated case (10-12/08/2016).

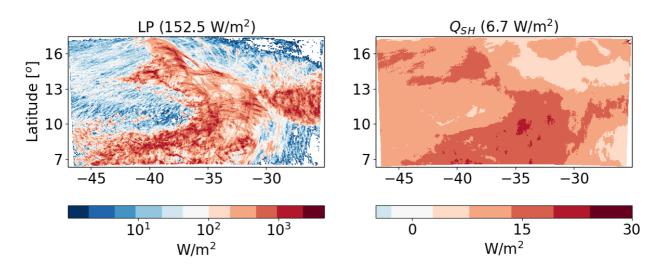




913 Figure 11. histograms of ICON simulated vertical velocity at the level of 500 mb <u>for a clean (CDNC = 20 cm⁻³)</u> 914 and polluted (CDNC = 200 cm⁻³) simulations (upper), and the time evolution of the net upwards water (liquid 915 and ice) mass flux (lower) <u>for the different CDNC simulations for a clean (CDNC = 20 cm⁻³) and polluted</u> 916 (CDNC = 200 cm⁻³) simulations for the shallow-cloud dominated case (10-12/08/2016). The 500 mb level is 917 chosen as it represents the transition between the warm part to the cold part of the clouds. <u>In the histogram</u> 918 only two simulations are presented for clarity.

920 Deep-cloud dominated case -16-18/08/2016

Next, we analyse the atmospheric energy budget for the deep-cloud dominated case (Fiona 921 tropical storm - Fig. 12). As opposed to the shallow-cloud dominated case, in this case the LP 922 contribution dominates over the radiative cooling and hence the energy imbalance-residual R is 923 positive and large, suggesting divergence of dry static energy out of the domain. This difference 924 in the base line atmospheric energy budget between the different cases simulated here, enable an 925 examination of the aerosol effect on the atmospheric energy budget under contrasting initial 926 927 conditions. As in the shallow-cloud dominated case, the Q_R values varies between small values (especially at the regions that were mostly covered by deep clouds) to larger negative values 928 (dominated at the regions that were coved by shallow clouds). The Q_{SH} is positive and an order 929 of magnitude smaller than the Q_R and LP, similar to the shallow-cloud dominated case. 930



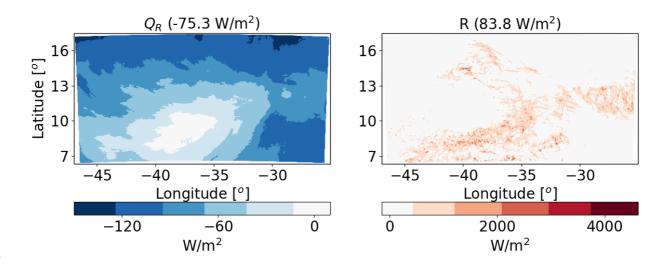
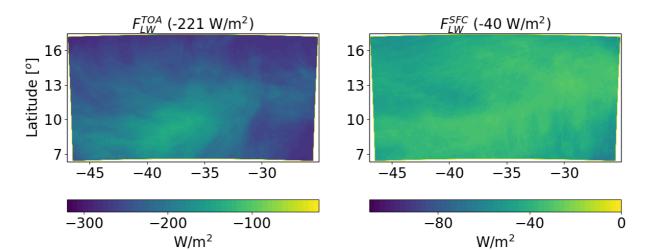


Figure 12. Spatial distribution of the time mean of the different terms of the energy budget for the ICON simulation of the deep-cloud dominated case (16-18/08/2016) with CDNC = 20 cm⁻³. The terms that appear here are: *LP* - latent heat by precipitation, Q_{SH} - sensible heat flux, Q_R - atmospheric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

936

Further examination of the radiative fluxes (Fig. 13) demonstrates again the resemblance in the 937 spatial structure between Q_R and F_{LW}^{TOA} . As compared to the shallow-cloud dominated case, since 938 the clouds are more opaque and cover larger fraction of the sky, there is a decrease in the 939 magnitude of all fluxes (in different amount). For example, F_{SW}^{SFC} is lower by 41 W/m² 940 (representing larger SW reflectance back to space) and the magnitude of F_{LW}^{TOA} by 47 W/m² as 941 compare to the shallow-cloud dominated case. The combined effect of the radiative flux 942 differences between the two cases is a decrease of the atmospheric radiative cooling by 39.6 943 944 W/m^2 (-114.7 compare with -75.3 W/m^2 – see Figs. 5 and 13).



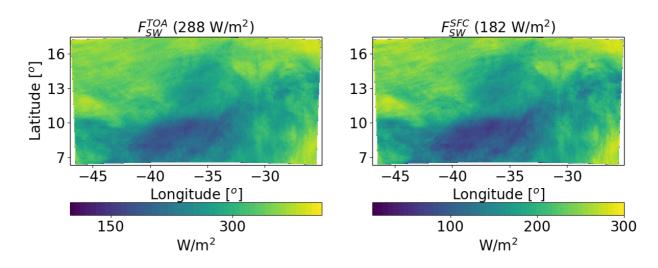


Figure 13. Spatial distribution of ICON simulated time-mean longwave (LW) and shortwave (SW) radiation
fluxes at the top of atmosphere (TOA) and surface (SFC) for a simulation of the deep-cloud dominated case
(16-18/08/2016) with CDNC = 20 cm⁻³. The domain and time mean value of each term appears in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

949

950 Response to aerosol perturbation – deep-cloud dominated case

For the deep-cloud dominated case, an increase in CDNC results in a decrease in *LP* by -0.3 W/m². Again, this difference is due to <u>a</u> 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 (Fig. 10.0 W/m² (Fig. 14) compared with 1.6 W/m² in the shallow-cloud dominated case (Fig. 6).

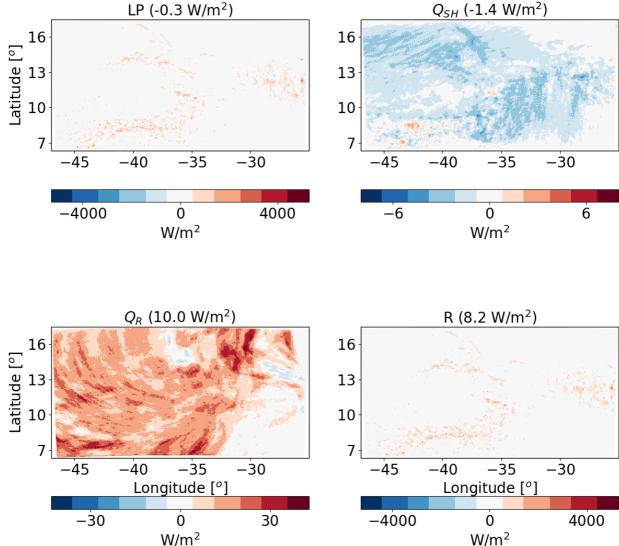


Figure 14. The differences between polluted (CDNC = 200 cm⁻³) and clean (CDNC = 20 cm⁻³) ICON simulations of the time-mean terms of the energy budget for the deep-cloud dominated case (16-18/08/2016). The terms that appears here are: *LP* - latent heat by precipitation, Q_{SH} - sensible heat flux, Q_R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

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/m² at TOA and -12.3 W/m² 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/m² atmospheric radiative cooling (which decrease some of the LW

warming). The net TOA total (SW+LW) radiative flux change is about -1.9 W/m². The trends in
the mean cloud properties (Figs. 16 and 17 below) can explain this large radiative response.

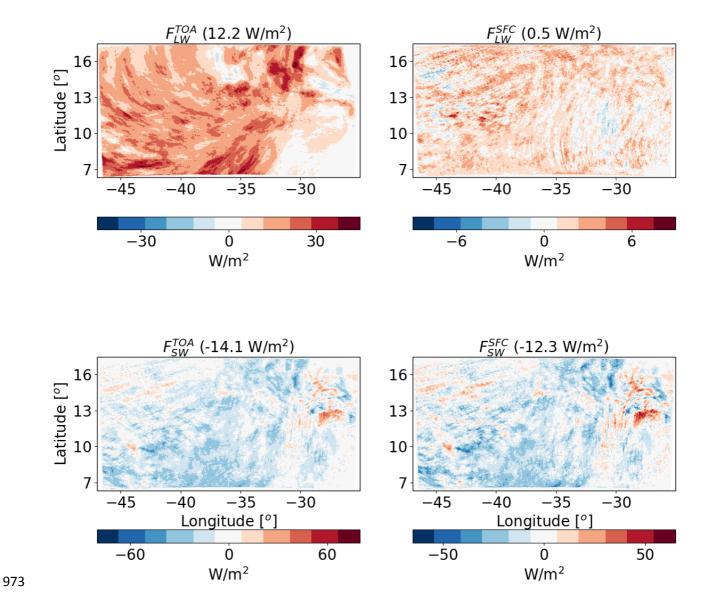


Figure 15. The differences between polluted (CDNC = 200 cm⁻³) and clean (CDNC = 20 cm⁻³) ICON
simulations of the time mean radiative longwave (LW) and shortwave (SW) fluxes at the top of atmosphere
(TOA) and surface (SFC) for the deep-cloud dominated case (16-18/08/2016). The domain and time mean
value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

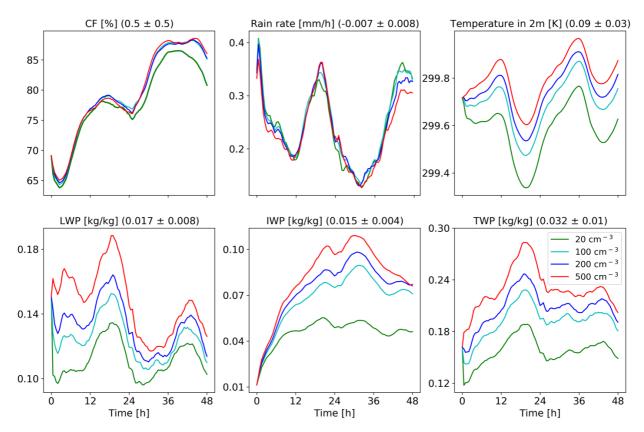
Figure 16 presents some of the domain mean properties as a function of time for the deep-cloud
dominated case. It demonstrates an increase in CF with CDNC which is more significant during
the second day of the simulation. This is opposite to the CF reduction in the shallow-cloud
dominated case (Fig. 8). It also demonstrates a very significant increase in LWP and, even more

(in relative terms), in IWP and thus also in TWP. The increase in CF and water content can 983 explain the decrease in SW fluxes both at TOA and surface (Fig. 15) as more SW is being 984 reflected back to space. The larger SW reflection under increased CDNC is also contributed to 985 by the Twomey effect (Twomey, 1977). Re-running the simulations without the Twomey effect 986 result in 9.6 W/m² reduction in the TOA SW flux as compare to 14.1 W/m² with the Twomey 987 effect on. We note that the relative role of the Twomey effect (compare to the cloud adjustments 988 989 - CF and TWP) is larger in the shallow-cloud dominated case as compared to the deep-cloud dominated case $(-14.19.6 \text{ W/m}^2 \text{ and } -9.614.1 \text{ W/m}^2 \text{ for simulations with and without the Twomey})$ 990 effect in the deep-cloud dominated case, compare to -1.77.5 W/m² and -1.77.5 W/m² in the 991 shallow-cloud dominated case, respectively). However, it should be noted that the Twomey 992 effect due to changes in the ice particles size distribution was not considered. In this case, unlike 993 in the shallow-cloud dominated case, the three contributions to the SW changes (CF, Twomey 994 and LWP/IWP, e.g. (Goren and Rosenfeld, 2014)) all contribute to the SW flux reduction (Fig. 995 15 presents the results of all contributors). Off-line sensitivity tests demonstrate that the relative 996 contribution of the TWP-water content and the CF to the increase in SW reflectance is roughly 997 998 $\frac{3}{4}$ and $\frac{1}{4}$, respectively.

999 The vertical profile changes with CDNC (Fig. 17) demonstrate a consistent picture of a decrease 1000 in CF in low clouds and a significant increase in CF and liquid and ice content at the mid and upper troposphere. The CF increase at the upper troposphere, and especially the increase in the 1001 ice content, can explain the decrease in the outgoing LW radiation (Fig. 15). The increase in ice 1002 content at the upper troposphere is in agreement with recent observational studies (Gryspeerdt et 1003 1004 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. 1005 19), demonstrates a substantial increase with the increase in CDNC (Chen et al., 2017), which 1006 1007 occurs due to the increase in the water content (Fig. 17) and the delay in the rain formation to 1008 higher levels (Heikenfeld et al., 2019), even without a large change in the vertical velocity or 1009 cloud fraction at this level (Fig.17) due to the increase in the water content (Fig. 17) and the delay in the rain formation to higher levels (Heikenfeld et al., 2019). Similar to the shallow-cloud 1010 dominated case (Fig. 8), the near surface temperature monotonically increases with CDNC, while 1011 1012 the effect on the mean rain rate is small.

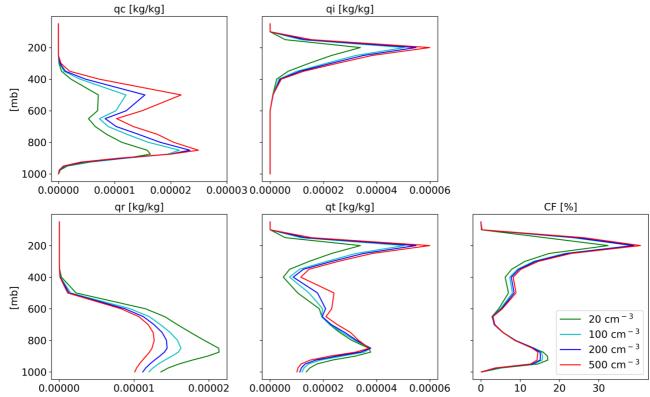
The differences in the thermodynamic evolution between polluted and clean conditions for this
case (Fig. 18), demonstrate the same trend as in the shallow-cloud dominated case (Fig. 10).
Here again, we note an increase in the humidity at the mid and upper troposphere, that contribute

to the reduction in the outgoing LW flux. The deepening, drying and warming of the boundary 1016 layer are observed in this case as well. Both the increase in humidity at the mid-upper troposphere 1017 and the deepening of the boundary layer (Seifert et al., 2015) could cause a reduction of the 1018 outgoing LW flux. To distinguished the effect of clouds and humidity at the different levels on 1019 the outgoing LW flux, we have conducted sensitivity off-line radiative transfer calculations using 1020 BUGSrad. As in the shallow-cloud dominated case, the difference in outgoing LW flux between 1021 clean and polluted conditions primarily emerges from the CDNC effect on clouds. The small 1022 remaining effect of the clear sky ($\sim 0.2 \text{ W/m}^2$) is contributed by the change in the humidity at the 1023 1024 mid and upper troposphere rather than by the deepening of the boundary layer (which would lead to LW emission from lower temperatures and is expected to be more significant under lower free 1025 troposphere humidity conditions). 1026





1028Figure 16. Domain average properties as a function of time for the different CDNC simulations for the deep-1029cloud dominated case. The properties that are presented here are: cloud fraction (CF), rain rate, temperature1030in 2 m, liquid water path (LWP - based on the cloud water mass, excluding the rain mass for consistency1031with satellite observations), ice water path (IWP) and total water path (TPW = LWP + IWP). For each1032property₂ the mean difference between all combinations of simulations, normalized to a factor 5 increase in1033CDNC, and its standard deviation appear in parenthesis.



1035 Figure 17. Domain and time average vertical profiles for the different CDNC simulations for the shallow-1037 cloud dominated case. The properties that are presented here are: cloud droplet mass mixing ratio (qc – for 1038 clouds' droplets with radius smaller than 40 μ m), ice mass mixing ratio (qi), rain mass mixing ratio (qr - for 1039 clouds' drops with radius larger than 40 μ m), total water mass mixing ratio (qt = qc+qi+qr), and cloud 1040 fraction (CF).

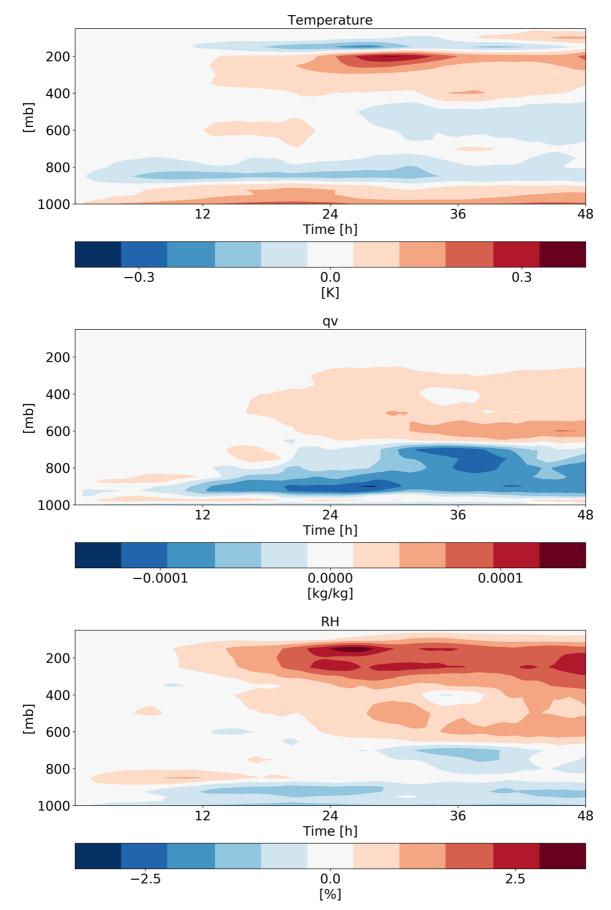
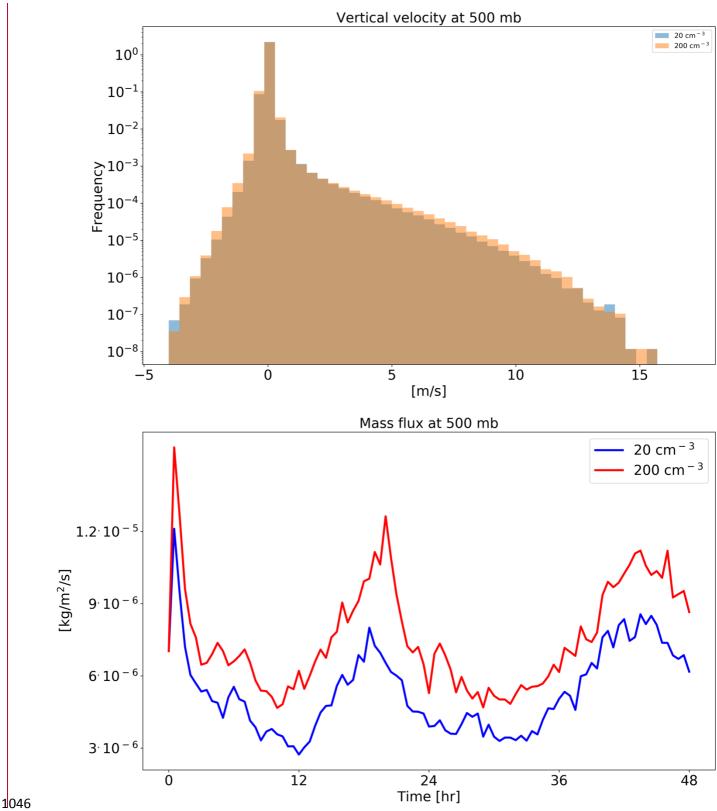
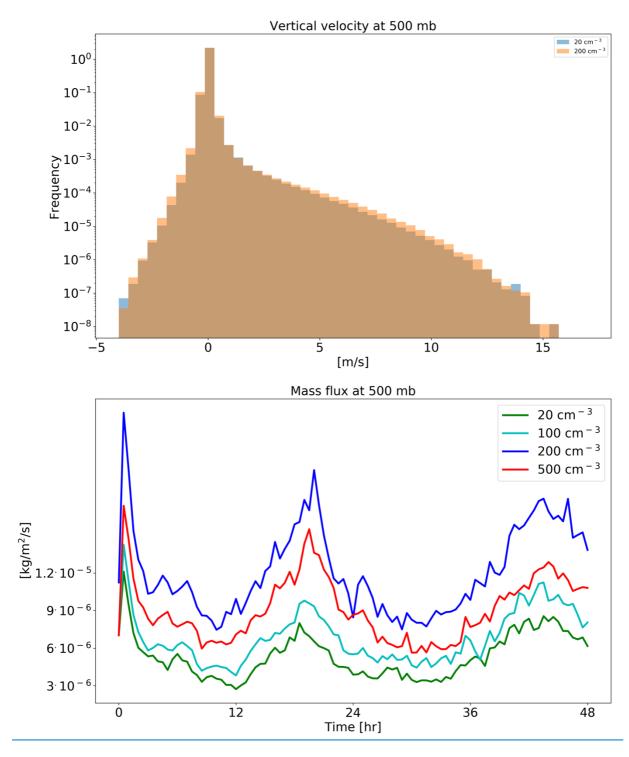


Figure 18. <u>Time-height</u> <u>Hoymöller</u> diagrams of the differences in the domain mean temperature, specific

- 1043 humidity (qv) and relative humidity (RH) vertical profiles between polluted (CDNC = 200 cm⁻³) and clean
- 1044 (CDNC = 20 cm^{-3}) simulations for the deep-cloud dominated case (16-18/08/2016).







1048Figure 19.- histograms of ICON simulated vertical velocity at the level of 500 mb for a clean (CDNC = 201049cm⁻³) and polluted (CDNC = 200 cm⁻³) simulations (upper), and the time evolution of the net upwards water1050(liquid and ice) mass flux (lower) for the different CDNC simulations for the deep-cloud dominated case (16-105118/08/2016). The 500 mb level is chosen as it represents the transition between the warm part to the cold part1052of the clouds. In the histogram only two simulations are presented for clarity. histograms of ICON simulated1053vertical velocity at the level of 500 mb (upper), and the time evolution of the net upwards water (liquid and1054iee) mass flux (lower) for a clean (CDNC = 20 cm⁻³) and polluted (CDNC = 200 cm⁻³) simulations for the deep-

1055cloud dominated case (16-18/08/2016). The 500 mb level is chosen as it represents the transition between the1056warm part to the cold part of the clouds.

1057

1058 Summary and conclusions

Two different case studies of tropical cloud systems over the Atlantic Ocean were simulated 1059 using the ICON numerical model in a cloud resolving configuration with 1.2 km resolution and 1060 a relatively large domain (~22° x 11°). The cases represent dates from the NARVAL 2 field 1061 campaign that took place during August 2016 and have different dominant cloud types and 1062 different dominating terms in their energy budget. The first case (10-12/8/2016) is shallow-cloud 1063 dominated and hence dominated by radiative cooling, while the second case (16-18/8/2016) is 1064 dominated by deep convective clouds and hence dominated by precipitation warming. The main 1065 objective of this study is to analyse the response of the atmospheric energy budget to changes in 1066 cloud droplet number concentration (CDNC), which serve as a proxy for (or idealized 1067 representation of) changes in aerosol concentration. This enables better understanding of the 1068 1069 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; 1070 1071 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 1072 can be significantly perturbed by changes in CDNC and that the magnitude of the effect is cloud 1073 1074 regime dependent (even for a given geographical region and given time of the year as the two 1075 cases are separated by less than a week).

Figure 20 summarizes the energy and radiation response of the two simulated cases to CDNC 1076 perturbations. It shows that the atmosphere in the deep-cloud dominated case experiences a very 1077 strong atmospheric warming due to an increase in CDNC (10.0 W/m²). Most of this warming is 1078 caused by a reduction in the outgoing LW radiation at the TOA. The SW radiative fluxes (both 1079 1080 at the TOA and surface) is also significantly modified but their net effect on the atmospheric column energy budget is small. The net TOA radiative fluxes change in this case is -1.9 W/m². 1081 Beside the atmospheric radiative warming, changes in precipitation (~-0.3 W/m²), and in sensible 1082 1083 heat flux (Q_{SH_2} -1.4 W/m²) also contribute to the total trend as a response of increase in CDNC. We note that since 1 mm/hr of rain is equivalent to 628 W/m², even negligible changes in 1084 1085 precipitation of less than 0.5 mm over 48 hr (as seen in our simulations) can still appear as 1086 significant changes in the atmospheric energy budget and contribute a few W/m².

The response of the radiative fluxes can be explained by the changes in the mean cloud and 1087 thermodynamic properties in the domain. The mean cloud fraction (CF) increases with the 1088 increase in CDNC (Fig. 16) while the vertical structure of it indicates a reduction in the low 1089 cloud fraction (below 800 mb) and an increase in the mid and upper troposphere CF (Fig. 17). 1090 The water content (both liquid and ice) also increase with the increase in CDNC (Figs. 16 and 1091 1092 17) with increasing amount with height. These changes in the mean cloud properties drive both the reduction in SW fluxes at TOA and surface and LW flux at TOA as the clouds become more 1093 opaque (Koren et al., 2010; Storelvmo et al., 2011) and cover a larger fraction of the sky. In 1094 1095 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 1096 CDNC, (due to increase mass flux to the upper troposphere) which further decreases the outgoing 1097 LW flux at the TOA. However, the vast majority of the LW effect emerges from the changes in 1098 1099 clouds.

1100 Both the increase in water vapor and ice content inat the upper troposphere are driven by an 1101 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 1102 1103 increase in the water mixing ratio inat the mid-troposphere rather than by increase in vertical velocity (Figs. 11 and 19) or in cloud fraction (Fig. 17). The ice content inat the upper 1104 troposphere is also increased due to reduction in the ice falling speed (Grabowski and Morrison, 1105 2016), while the increased relative humidity at these levels, further increases the ice particle 1106 lifetime due to slower evaporation. However, the increase in water mass flux to the upper layers 1107 1108 is not accompanied with an increase in precipitation as predicted by the classical "invigoration" paradigm (Altaratz et al., 2014; Rosenfeld et al., 2008), which suggest that some compensating 1109 mechanisms are operating (Stevens and Feingold, 2009). 1110

In the shallow-cloud dominated case (which also contains a significant amount of deep 1111 convection), the response of Q_R is weaker but still substantial (a total decrease in the atmospheric 1112 1113 radiative cooling of 1.6 W/m² - Fig. 20). The weaker total response under the shallow-cloud dominated conditions is due to the smaller role of the ice part in this case. Here again, the changes 1114 in Q_{SH} decrease about -1.4 W/m² of this atmospheric warming. As in the deep-cloud dominated 1115 case, most of the atmospheric radiative warming is caused by reduction in the outgoing LW flux, 1116 while the surface and TOA SW fluxes changes are non-negligible but cancel each other out (in 1117 terms of the atmospheric energy budget – reflecting small SW atmospheric absorption changes). 1118 However, a significant TOA net (SW+LW) radiative flux change of ~-5.2 W/m² remains. In this 1119

case, the cloud-mean effect on radiation is more complicated. While CF decreases with 1120 increasing CDNC, the mean water path (both LWP and IWP) increases (Fig. 8). As in the deep-1121 cloud dominated case, the increase in the water content occurs mostly at the mid and upper 1122 troposphere, while the decrease in CF occurs mostly in the lower troposphere (Fig. 9). In terms 1123 of the SW fluxes, the effect of the decrease in low CF (decrease SW reflections) and the increase 1124 1125 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 1126 1127 reflected back to space at TOA and a net negative flux change (including also the LW).

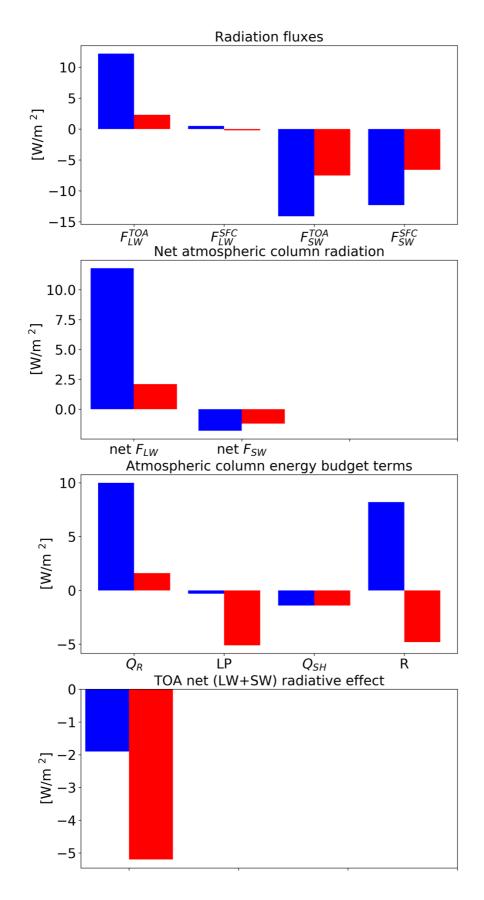
1128 There exists a large spread in estimates of aerosol effects on clouds for different cloud types and different environmental conditions. In this study, as we use a relatively large domain (22° x 11°) 1129 1130 and two different dates (each for two days), we sample many different local environmental conditions and cloud types. Such more realistic setups (although with lower spatial resolution) 1131 1132 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 1133 1134 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 1135 1136 conclusions over a wider range of initial conditions, we have conducted a large ensemble of 1137 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 1138 1139 conclusions presented in this paper are robust and hold also for a wide range of initial conditions 1140 representative for this area. In addition, the realistic setup with the continuously changing boundary conditions and systems that pass through the domain, which are used here, prevent 1141 conclusions that might be valid only in cyclic double periodic large eddy simulations, as the 1142 background meteorological conditions change more realistically (Dagan et al., 2018b). Another 1143 1144 uncertainty in the assessment of the aerosol response are the large differences between different 1145 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 1146 1147 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 1148 1149 include the temporal evolution of the aerosol concentration. Feedbacks between the aerosol concentration and clouds processes (such as wet scavenging), as well as the direct effects of 1150 aerosol on radiation would add another layer of complexity that should be accounted for in future 1151 1152 work.

Generally, the global mean aerosol radiative forcing is estimated to be negative (Boucher et al., 1153 2013; Bellouin et al., 2019). However, these global aerosol forcing estimates have so far not 1154 included the radiative forcing associated with potential effects of aerosols on deep convection -1155 and these effects are not represented in most current climate models due to limitations in 1156 convection parameterisations, with only a few exceptions (Kipling et al., 2017; Labbouz et al., 1157 2018). Here we demonstrate the existence of non-negligible aerosol radiative effects (of -5.2 and 1158 -1.9 W/m² for the shallow and deep cloud dominated cases, respectively) in tropical cloud 1159 systems, that contained both deep and shallow convective clouds, with significant SW and LW 1160 1161 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 1162 regime the effect may vary with the meteorological conditions. 1163

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

1166 increased convergence in shallow clouds regions, can have effects on the large-scale circulation.

1167 This should be investigated in future work.





1169 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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