Response to the reviewers' comments on

Ensemble daily simulations for elucidating cloud-aerosol interactions under a large spread of realistic environmental conditions

We would like to thank the reviewers for their constructive and thoughtful comments that helped us to improve our paper.
Please find a point by point reply to all of the reviewers' comments (in blue) below.

Reviewer #1:

This study performs a sensitivity test to the CCN concentrations in a domain of 3x3 degrees just to the west of Barbados. It runs two full months of actual weather, one in December 2013 and one in August 2016. A major conclusion that is well supported by the study is the variability of the indicated aerosol effects on different days, and the implication that conclusions from single case studies should not be generalized for a large range if situations.
The rest of the quantitative conclusions of the study with respect to aerosol effects are limited by the fidelity of the model that was used and to the way of its application. The model that was used is a two-moment bulk microphysical scheme (Seifert and Beheng, 2006b). The model has severe major limitations:

Reply: Before replying to each point, we would like to start with a general remark. The main concern of the reviewer is the use of a two-moment bulk microphysical scheme as opposed to a bin scheme. We agree with the reviewer that bin schemes can have advantages in representing some of the aerosol effect on clouds, and as the reviewer mentions below, the first author of this study worked quite a bit with that type of microphysical schemes previously (i.e. (Dagan et al., 2015a;Dagan et al., 2015b, 2018;Dagan et al., 2017;Dagan et al., 2016;Heiblum et al., 2016). Hence, we fully appreciate and are aware of the limitations of our model. However, one should also remember that bin microphysical schemes have their own limitations such as broadening of the droplet size distribution due to numerical diffusion (Morrison et al., 2018). In addition, different bin microphysical schemes demonstrate a high inter-model spread in predicting warm rain production (VanZanten et al., 2011, Adrian Hill, personal communication) as well as a large spread in predicting ice processes (Grabowski et al., 2019; Xue et al., 2017). This large uncertainty in microphysics schemes – of all kinds – is also supported by ongoing work in the deep convection case study of the Aerosol, Clouds, Precipitation and Climate (ACPC) initiative (van den Heever et al., in prep.). Overall, limited observations leave the representation of mixed-phase cloud microphysics poorly constrained. More fundamentally, it is not a given (and often not true) that more complex model representations, such as bin microphysics schemes in this case, will by default evaluate better against observations than simpler representations. Instead, we would argue that the field has not yet reached a consensus about the optimal representation of cloud microphysics (Grabowski et al., 2019) and similar arguments as made in this review could be about the use of bin schemes versus fully lagrangian microphsics or superdroplet schemes.

In addition to the ongoing debate in our field about bin versus bulk microphysical approaches, there is the clear issue of computational expense which limits many bin-model studies to small domains, coarser resolution or shorter simulations (Khain et al., 2015). Addressing the representativeness of such simulations is a particular concern of our work: the main aim of our current study is to examine aerosol effect under a *wide range of initial conditions*, which requires numerus simulations. To do so we simulate 248 day-long simulations (62 days times two CDNC conditions per day times two simulations for each case to estimate the Twomey effect). Our simulations are conducted on relatively large domain of ~300 x 300 km (compared with many previous studies) with relatively high vertical and horizontal resolutions (75 vertical levels and 1.2 km horizontal resolution). This large amount of simulations required a huge amount of computational time. Using a bin microphysical scheme would enlarge the computation time by 1-2 orders of magnitude. Hence, this is impossible for us to conduct at this stage and we do not believe that it would affect our main conclusions.

For studying cloud-aerosol interactions using numerical simulation one must make some simulation choices. Currently, it is impossible to simulate cloud-aerosol interactions with the most detailed microphysical scheme (if we knew the optimal approach), with the highest spatial and temporal resolution, a large domain and a large ensemble of initial conditions. We must make a choice. In this study, we choose to focus on the effect of the initial conditions and examine a wide range of them (which is the novelty of this study). Other studies, e.g. the ACPC deep convection case study, will focus on convective cloud microphysics. As will be explained in more detail below, even though we are aware of the limitation of our simulation choices, we still think (and the reviewer seems to agree) that the main conclusion of our paper (citing from the reviewer above "…the variability of the indicated aerosol effects on different days, and the implication that conclusions from single case studies should not be generalized for a large range if situations") is "well supported".

1. The model assumes saturation adjustment, thus cannot realize the invigoration mechanism that is incurred by the warm cloud invigoration mechanism mediated by the aerosol control on the supersaturation that was co-authored by the first author of this study (Koren et al., 2014). In that paper it is shown that the lack of supersaturation adjustment is responsible for most of the substantial invigoration of water convective clouds on the background of very low CCN.

Reply: Thank you for this comment. As the reviewer mentioned we are well aware of the limitation of using saturation adjustment and worked quite a bit on this topic previously. However, please note that in our simulations we use a temporal resolution of 12 sec. The phase change relaxation time of condensation and evaporation is usually on the order of a few seconds, and even under extremally clean conditions is not more than 10 sec (Pinsky et al., 2013). Hence, even if we would use a microphysical scheme that explicitly resolves condensation and evaporation, the humidity is expected to get back to saturation on shorter time scales then the temporal resolution of the model, and hence, practically we will be in "saturation adjustment" conditions anyway.

Following this comment, we added clarifications to the revised manuscript:

"In addition, we note that use of a microphysical scheme which assumes saturation adjustment reduces the sensitivity of the clouds to some of the aerosol effect (Koren et al., 2014; Dagan et al., 2015a; Heiblum et al., 2016; Fan et al., 2018)."

"In addition, using a microphysical scheme that assumes saturation adjustment reduces the sensitivity of the clouds to aerosol perturbation (Koren et al., 2014; Dagan et al., 2015a; Heiblum et al., 2016; Fan et al., 2018). However, this might be a small effect in our case as the phase change relaxation time of condensation and evaporation is usually on the order of a few seconds (Pinsky et al., 2013). Hence, even if we would use a microphysical scheme that explicitly resolves condensation and evaporation, the humidity is expected to get back to saturation on shorter time scales then the temporal resolution of the model (12 sec), and hence, practically we will be in "saturation adjustment" conditions anyway."

2. The mechanism of convective invigoration mediated by aerosol control of the super-saturation becomes even more important in deep convective clouds (Fan et al., 2018). Therefore, selecting a model with saturation adjustment misses most of the aerosol effect on deep convective clouds.

Reply: As was mentioned above, one must make some simulation choices and, in this case, in order to sample a large spread of initial conditions, we choose to use a temporal resolution which will anyway simulate conditions close to saturation adjustment. Other studies will rightfully focus on the representation of microphysics. In addition, whether or not saturation adjustment is responsible for *most* of the aerosol effect on deep convective clouds is still to be determined.

In the revised manuscript we acknowledge that our simulations do not include the above-mentioned effect and we cite the paper the reviewer mentioned (see above).

3. Furthermore, in the model, droplet nucleation does not change the CCN spectrum, as acknowledged in the manuscript. But the scavenging of aerosol by precipitation serves as a strong positive feedback to amplify the difference in the aerosol effect between raining and not raining clouds.

Reply: Thank you for this comment. The scavenging of aerosol by precipitation could also serve as a "buffering" mechanism and reduce the difference between clean and polluted conditions. For example, if due to cloud invigoration mechanisms, initially there would be stronger rain rates under polluted conditions (Fan et al., 2013), and hence more scavenging, this could clean the atmosphere and bring it closer to the low aerosol case. However, in marine stratocumulus clouds it was shown, as the reviewer mentioned, that scavenging could amplify the difference between clean precipitating conditions and polluted non-precipitating conditions (Koren and Feingold, 2011).

The lack of inclusion of the scavenging mechanism and the feedback involved is mentioned in the manuscript:

"Using fixed CDNC avoids the uncertainties involved in the representation of aerosol processes in numerical models (Rothenberg et al., 2018), however, it limits potential feedbacks between clouds and aerosols, such as through aerosol scavenging (Yamaguchi et al., 2017)."

4. In addition, the 2M scheme does not suppress rain in high CCN concentrations to the extent that occurs in reality, where rain is suppressed pretty much when cloud drop effective radius is smaller than 14 micrometer (Chen et al., 2008; Freud et al., 2012; Gerber, 1996; Prabha et al., 2011; Rosenfeld et al., 2012; Van Zanten et al., 2005).

Reply: No model is perfect. Bin microphysical schemes have their own problems in predicting rain such as artificial broadening of the droplet size distribution due to numerical diffusion (Morrison et al., 2018) and a too early initialization of rain due to that. In addition, different bin microphysical schemes do not converge on the warm rain production rates (VanZanten et al., 2011; Adrian Hill, personal communication). As was cited above, the limitations of our simulations are now better explained in the manuscript.

5. The model misses the processes which can lead to positive net TOA warming due to aerosols, as simulated using SBM by Fan et al. (2012).

Reply: Thank you. In a recent study (Dagan et al., 2019), using the same model used here, we showed that aerosol could generate net atmospheric column warming (up to 10 Wm^{-2}) due to a reduction in outgoing longwave flux at TOA. This trend is driven by an increase in ice content and cover at the upper troposphere, a similar mechanism as described in Fan et al. (2012). Whether or not this reduction in longwave flux at TOA caused a positive net TOA warming (overcome the shortwave effect) is still an open question and depend on the environmental conditions (Koren et al., 2010). However, from figure 10 in the main text we can see that this does happen using our model on some of the summer days. This is already mentioned in the text:

"However, during the summer month, the mean effect is less negative (-1.7 W/m^2) and for some of the days it could even be positive due to the effect of the CDNC on the ice water content (Fig. 5 and Table 2)."

In addition, the paper the reviewer pointed at (Fan et al., 2012) was added to the revised manuscript:

"It was shown that the total column atmospheric radiative warming ($Q_R = (F_{SW}^{TOA} - F_{SW}^{SFC}) + (F_{LW}^{TOA} - F_{LW}^{SFC})$, defined as the rate of net atmospheric diabatic warming due to radiative shortwave (SW) and longwave (LW) fluxes at the surface (SFC) and top of the atmosphere (TOA), when all fluxes positive downwards), is substantially increased with CDNC in a deep-cloud dominated case (by ~10 W/m^2), while a much smaller increase (~1.6 W/m^2) is shown in a shallow-cloud dominated case. This trend is caused by an increase in the upward mass flux of ice and water vapor to the upper troposphere that leads to reduced outgoing longwave radiation (Fan et al., 2012)."

6. The model resolution of 1200 m is insufficient to resolve properly the trade wind cumulus.

Reply: As was stated above, in order to sample a large range of initial conditions and cloud regimes we had to make some simulations choices. However, we still believe that our set-up captures the main processes acting on cloud-aerosol interaction (Naumann and Kiemle, 2019). We also compare our model results with ground-base measurements (Figs. S1 and S2, SI),

which demonstrates that the model does a reasonable job (see also a resent published research examining the effect of model resolution, using the same model and the same region, by Naumann and Kiemle (2019), demonstrating a "good skill" by the model compared to observations at a similar resolution).
The limitation of the horizontal resolution is now also mentioned in the revised manuscript:
"We also note that using 1200 m horizontal resolution does not properly resolve all shallow cumulus clouds (Naumann and Kiemle, 2019)."

7. The simulation is allowed 12 hours for spin up time. This also means that clouds in air mass that enter the border of the domain (typically from the east) require that much time to spin up. But the whole domain of 3 degrees from east to west is 325 km, divided by 12 hours equals 27 km/hour or 7.5 m/s. This means that when air mass speed is larger than that, the spin up would not be reached throughout the domain. The actual mean surface wind at Barbados airport is easterly 14 knots, which is 7.2 m/s, with little variation between winter and summer. Therefore, most of the simulated clouds are well within the spin-up time.
Reply: Thank you. The 12 hours spin-up time which is mentioned in the manuscript is the spin-up time required when starting the simulations from (ECMWF) reanalysis data. In our case we start our simulations from ICON simulations with a similar resolution (Klocke et al., 2017). The only main difference between the simulations of Klocke et al, (2017) and our simulations is the microphysical scheme. Hence, the spin-up time required is only few 10's of minutes (occurs within the cloud lifetime). An evidence for that can be seen in Figs. 8 and 16 in Dagan et al. (2019), which present time series of the domain mean properties and show that its responses to the microphysical perturbation within the first 30 min.
This is now better explained in the revised manuscript:
"Each simulation is conducted for 24 hours, starting from 12 UTC - 12 hours after the original simulations of Klocke et al., 2017 were initialized from reanalysis data, to reduce spin-up effects. Using initial and boundary conditions based on ICON simulations with similar resolution, as in Klocke et al. (2017), reduces the spin-up effects."

All these problems lend very little credibility to the conclusions with respect to the quantitative aerosol effects on the clouds. Presently SBM simulations are possible for the domain of this study, although quite more expensive. The fact that bulk models run faster is no longer a justification to use them for evaluating aerosol microphysical effects without addressing these issues.

Reply: As was mentioned above, using SBM is not possible for simulating 248 day-long, 300km by 300km domain simulations, which are needed for examining the dependency of the aerosol effect on the initial conditions in a robust manner. In addition, we note that there exists no consensus about the general preferability of SBM over bulk schemes based on observational evidence. We disagree with the implicit, yet unproven, assumption that only simulations based on SBM are credible, which would invalidate a large part of the existing literature.

In summary, I would recommend publication only after all these caveats will be explicitly highlighted, and the conclusions of the paper will clearly take them into account.

What is the point to run very fast with 2-Moment bulk scheme when running in the wrong direction?

Reply: As was mentioned above, it is far from being a consensus in our field that two-moment bulk schemes are running in the wrong direction – but that is not the focus of the presented work. However, we are aware of the limitations of cloud microphysics schemes and have highlighted this in the revised manuscript and the conclusions.

Reviewer #2:

This study is a follow-up to a paper exploring two case studies from the NARVAL campaign that uses ensemble simulations of two months (of which the original cases were a subset) to analyze the robustness of inferences regarding aerosol-cloud interactions that can be made on the basis of a small number of cases. Certain changes (such as in shortwave reflections and boundary layer deepening that lowers lower tropospheric stability) appear robust whereas others (such as cloud fraction and precipitation changes) appear less so. Seasonal differences in response can be explained via different responses in different cloud regimes, particularly due to ice-phase effects in deep clouds during the summer.

The manuscript is in very good shape and only requires some very minor revisions, in my estimation. If not for the comment below regarding the reasonableness of the "lower bound" language, I'd be happy to accept as is.

Reply: We would like to thank the reviser again for the constructive review and we are happy by the assessment that our paper is in very good shape.

Specific comments:

Page 2, Line 45: "As the anthropogenic activity. . ." is phrased somewhat awkwardly. Perhaps you can simplify to something like "Anthropogenic aerosol emissions may thus perturb Earth's radiation budget both directly by scattering and absorbing light and also indirectly through these cloud-mediated mechanisms."

Reply: Thank you for this suggestion. This sentence was changed according to the reviewer suggestion.

Page 4, Line 116: I'm glad you address this point. However, did you mean "interactions" or "feedbacks" instead of "involve"? Also, a relevant citation for the aerosol scavenging idea: Yamaguchi, T., Feingold, G., & Kazil, J. (2017). Stratocumulus to Cumulus Transition by Drizzle. Journal of Advances in Modeling Earth Systems, 9(6), 2333-2349. doi:10.1002/2017MS001104

Reply: Thank you. This sentence was changed and the suggested reference was added:

"Using fixed CDNC avoids the uncertainties involved in the representation of aerosol processes in numerical models (Rothenberg et al., 2018), however, it limits potential feedbacks between clouds and aerosols, such as through aerosol scavenging (Yamaguchi et al., 2017)."

Page 5, Line 138: I'm not convinced this is a reasonable lower bound, given that the relatively small domain size with fixed boundary conditions (which you argue would lead to an underestimate of aerosol effects) is not the only potential source of error, or necessarily the largest. I'd either like to see a fuller explanation of why the estimates should be seen as true lower bounds or a weaker statement simply explaining this particular source of error would tend to underestimate the effect compared to a simulation with a larger domain.

Reply: Thank you for this comment that helped us clarify this point. We agree with the reviewer that this statement was too strong in the previous version and hence in the revised manuscript we removed it:

"We note that although a 3^{o} x 3^{o} domain is larger than the domains used in many previous studies, it is still possible that the use of fixed boundary conditions for the different simulations under different CDNC conditions reduces some of the sensitivity as compared to simulations with larger domains such as in Dagan et al. (2019) (22^{o} x 11^{o})."

Page 6, Figure 2: It would be helpful if "LP" were defined somewhere in the text in addition to in the figure captions.

Reply: added.

"This demonstrates that the lower tropospheric stability (LTS), top of atmosphere shortwave flux (F_{SW}^{TOA}), and the atmospheric column radiative term (Q_R) are different in a statistically significant manner (p-value < 0.05) between the two different months. The differences in other parameters (cloud fraction – CF, liquid water path - LWP, ice water path – IWP, latent heat of precipitation – LP, and top of atmosphere longwave flux - F_{LW}^{TOA}) are not statistically significant (Table 1)."

Page 9, Line 226: I would add "likely" between "would" and "further" given that ice-phase microphysical changes can be quite complex.

Reply: added.

"Accounting for this effect would likely further increase the relative role of the Twomey effect compared to the cloud adjustment effects (CF and LWP/IWP adjustments)."

Page 11, Line 275: How significant is 12 versus 8 in this context? Is there any way to quantify the variability we could expect in deep-cloud days due to chance?

Reply: Thank you for this comment. It is indeed hard to say whether or not a 50% increase in the occurrence of deep convection between the two months (12 versus 8 days) is significant based only on this data. However, the difference between the summer and winter in the occurrence of deep convection in the Barbados region is well known (Stevens et al., 2016). In addition, in our data we do see a significant difference in the LTS between the two months (Fig. 2 and table 1 in the main text), which is consistent with the increase in deep convection occurrence.

Based on this comment we have added clarification to the revised manuscript:

"The separation into different cloud regimes also demonstrates that more deep-cloud days are occurring during the summer month as compared to the winter month (12 compare to 8) and that the deep clouds during summer are deeper and contain more water. The larger occurrence of deep convection during the summer month is consistent with the statistically significant reduction in LTS (Fig. 2 and Table 1) and is expected based on the local seasonality (Stevens et al., 2016)."

References: There are some typos and weird formatting issues with some references. A quick proofread should sort most of those out.

Reply: Thank you. The manuscript (including the references section) went thought proofreading.

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Ensemble daily simulations for elucidating cloud-aerosol interactions under a large spread of realistic environmental conditions

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Abstract

Aerosol effects on cloud properties and the atmospheric energy and radiation budgets are studied through ensemble simulations over two month-long periods during the NARVAL campaigns (December 2013 and August 2016). For each day, two simulations are conducted with low and high cloud droplet number concentrations (CDNC), representing low and high aerosol concentrations, respectively. This large data-set, which is based on a large spread of co-varying realistic initial conditions, enables robust identification of the effect of CDNC changes on cloud properties. We show that increases in CDNC drive a reduction in the top of atmosphere (TOA) net shortwave flux (more reflection) and a decrease in the lower tropospheric stability for all cases examined, while the TOA longwave flux and the liquid and ice water path changes are generally positive. However, changes in cloud fraction or precipitation, that could appear significant for a given day, are not as robustly affected, and, at least for the summer month, are not statistically distinguishable from zero. These results highlight the need for using large statistics of initial conditions for cloud-aerosol studies for identifying the significance of the response. In addition, we demonstrate the dependence of the aerosol effects on the season, as it is shown that the TOA net radiative effect is doubled during the winter month as compared to the summer month. By separating the simulations into different dominant cloud regimes, we show that the difference between the different months emerge due to the compensation of the longwave effect induced by an increase in ice content as compared to the shortwave effect of the liquid clouds. The CDNC effect on the longwave is stronger in the summer as the clouds are deeper and the atmosphere is more unstable.

Introduction

Cloud droplets form on suitable aerosols which can serve as cloud condensation nuclei. Thus, for vertical velocities which are sufficient to sustain aerosol activation, cloud droplet number concentration (CDNC) increases with increasing aerosol concentrations. Concomitantly with the increase in the CDNC, and assuming constant liquid water content, the initial cloud hydrometeor (liquid and ice particles) size distribution shifts to smaller sizes and becomes narrower, which may modulate cloud micro- and macro-physical properties (Khain et al., 2005;Koren et al., 2005;Heikenfeld et al., 2019;Chen et al., 2017;Altaratz et al., 2014;Seifert and Beheng, 2006a;Koren et al., 2014;Dagan et al., 2017;Dagan et al., 2018b), the rain production (Levin and Cotton, 2009;Albrecht, 1989;Tao et al., 2012;Dagan et al., 2015b) and the clouds' radiative effect (Koren et al., 2010;Storelvmo et al., 2011;Twomey, 1977;Albrecht, 1989). Anthropogenic aerosol emissions may thus perturb Earth's radiation budget both directly by scattering and absorbing light and also indirectly through these cloud-mediated mechanismsAs the anthropogenic activity involves aerosol emissions and aerosols may influence cloud radiative effects, the anthropogenic activity may perturb the Earth's radiation budget by this pathway. However, despite decades of effort of trying to better understand the processes involved, cloud-aerosol interactions are still considered one of the most uncertain anthropogenic effects on climate (Boucher et al., 2013).

The aerosol effect on clouds was previously shown to be 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;Glassmeier and Lohmann, 2016;Gryspeerdt and Stier, 2012;Christensen et al., 2016). In addition, even for a given cloud regime, small changes in the meteorological conditions may change the sign and magnitude of the aerosol effect (Dagan et al., 2015b;Fan et al., 2009;Fan et al., 2007;Kalina et al., 2014;Khain et al., 2008;Liu et al., 2019).

The fact that the aerosol effect on clouds and precipitation is dependent on the cloud regime and meteorological conditions, makes the quantification of its global effect challenging and uncertain (Mülmenstädt and Feingold, 2018;Bellouin et al., 2019). One way to overcome this challenge is by examining the aerosol effect for an ensemble of realistic co-varying initial conditions (as opposed to perturbing each environmental condition separately). This can be done by conducting ensemble/routine numerical simulations (such as those conducted in previous studies (Gustafson Jr and Vogelmann, 2015;Gustafson et al., 2017;Klocke et al., 2017)) focusing on aerosol effects. This methodology enables identifying, using large statistics, clouds and radiative properties that respond in a consistent manner to aerosol (noting that in a

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single-case studies some of the differences between different simulations could be just due to different realizations of the model (Grabowski, 2015)). This methodology also enables investigation of the aerosol effect on cloud and precipitation as a function of the initial conditions.

In a recent paper, focusing on two specific cases (each one for two days) and a relatively large domain (22^o x 11^o), the physical processes controlling the aerosol effect on the atmospheric energy budget were investigated (Dagan et al., 2019). It was shown that the total column atmospheric radiative warming (Q_R = $(\boldsymbol{F_{SW}^{TOA}} - \boldsymbol{F_{SW}^{SFC}}) + (\boldsymbol{F_{LW}^{TOA}} - \boldsymbol{F_{LW}^{SFC}})$, defined as the rate of net atmospheric diabatic warming due to radiative shortwave (SW) and longwave (LW) fluxes at the surface (SFC) and top of the atmosphere (TOA), when all fluxes positive downwards), is substantially increased with CDNC in a deep-cloud dominated case (by ~10 W/m^2), while a much smaller increase (~1.6 W/m^2) is shown in a shallow-cloud dominated case. This trend is caused by an increase in the upward mass flux of ice and water vapor to the upper troposphere that leads to reduced outgoing longwave radiation (Fan et al., 2012). The increase in mass flux is caused partially by an increase in vertical velocities (Koren et al., 2005;Rosenfeld et al., 2008;Dagan et al., 2018a) and mostly by an increase in the water content at the mid-troposphere (due to warm rain suppression) that increases the upward mass flux, even for a give vertical velocity. The change in net radiative fluxes at the TOA ($\boldsymbol{F_{SW+LW}^{TOA}}$) was shown to be -5.2 W/m^2 for the shallow-cloud dominated case and -1.9 W/m^2 for the deep-cloud dominated case. Dagan et al. (2019) also show that the cloud fraction responds in opposite ways to CDNC perturbations in the different cases, increasing in the deep-cloud dominated case and decreasing in the shallow-cloud dominated case. However, it is unclear how representative these results are as they are based on two specific cases. The ensemble simulations presented in this study could be used to examine the robustness of these aerosol effects using large statistics.

The focus of this study is on clouds over the Atlantic Ocean near Barbados (Fig. 1). Barbados is located north of the mean intertropical convergence zone (ITCZ) location, in a way that samples both the trade region, dominated by shallow cumulus during the boreal winter, and the transition to deep convection as the ITCZ migrates northward during boreal summer (Stevens et al., 2016). Hence, this location enables investigation of different cloud regimes and different meteorological conditions. In addition, the clouds near Barbados have been shown to be representative of clouds across the trade region (Medeiros and Nuijens, 2016).

Methodology

Ensemble daily simulations using the icosahedral nonhydrostatic (ICON) atmospheric model (Zängl et al., 2015) in a limited area configuration are conducted. ICON's dynamical core has been validated against several idealized cases as well as against numerical weather prediction skill scores (Zängl et al., 2015). The domain is located east of Barbados island and covers ~3^{o} x 3^{o} (Fig. 1). The simulations are aligned with the NARVAL (Next-generation Aircraft Remote-Sensing for Validation Studies (Klepp et al., 2014;Stevens et al., 2019;Stevens et al., 2016)) campaigns which took place during December 2013 (NARVAL 1) and August 2016 (NARVAL 2) in the northern tropical Atlantic. We use existing NARVAL convection-permitting simulations (Klocke et al., 2017) as initial and boundary conditions for our simulations and a two-moment bulk microphysical scheme (Seifert and Beheng, 2006b). For each day during these two months, two different simulations are started with identical initial conditions with different CDNC of 20 cm^{-3} (clean) and 200 cm^{-3} (polluted), resulting in an ensemble of 124 simulations. The different CDNC scenarios serve as proxy for different aerosol concentration conditions and are chosen as they represent the range typically observed over the ocean (Rosenfeld et al., 2019;Gryspeerdt et al., 2019). Using a fixed CDNC avoid the uncertainties involved in the representation of the aerosols processes in numerical models (Rothenberg et al., 2018), however, it limits potential feedbacks between clouds and aerosols, such as through involve with aerosol scavenging (Yamaguchi et al., 2017). In addition, we note that using microphysical scheme which assumes saturation adjustment reduces the sensitivity of the clouds to some of the aerosol effect (Koren et al., 2014; Dagan et al., 2015a; Heiblum et al., 2016; Fan et al., 2018).

Each simulation is conducted for 24 hours starting from 12 UTC - (12 hours after the original simulations of Klocke et al., 2017 started were initialized from reanalysis data, to reduce spin-up effects). Using initial and boundary conditions based on ICON simulations with similar resolution, as in Klocke et al. (2017), reduces the spin-up effects. The horizontal resolution is set to 1200 m and 75 vertical levels are used. The temporal resolution is 12 seconds and the output interval is 30 minutes. Interactive radiation is calculated every 12 minutes using the RRTM-G scheme (Clough et al., 2005;Iacono et al., 2008;Mlawer et al., 1997). The simulations include an interactive surface flux scheme and a fixed (for each day) sea surface temperature. As in Dagan et al. (2019), the simulations include representation of the Twomey effect, calculated with diagnosed cloud droplet effective radii from the microphysical scheme (Twomey, 1977). However, due to the large uncertainty involved in the ice microphysics and morphology, no Twomey effect due to changes in the ice particles size distribution was considered.

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In addition, the domain is setup to include the Barbados Cloud Observatory (BCO, (Stevens et al., 2016)) while minimising the island effect of Barbados (most of the domain is east of the island and only the east part of the island, which includes the BCO (13°N, 59°W), is included in the domain). Observations from the BCO are used for model evaluation (Figs. S1 and S2, supporting information), and demonstrate that the model performs well for low surface-SW-flux days but underestimates the flux for high-SW-flux days (usually under low cloud fraction). We note that although a 3 º x 3º domain is larger than the domains used in many previous studies, it is still possible that the use of fixed boundary conditions for the different simulations under different CDNC conditions reduces some of the sensitivity as compared to simulations with larger domains such as in Dagan et al. (2019) (22 º x 11º). Hence, the aerosol response we present here is estimated as the lower bound.

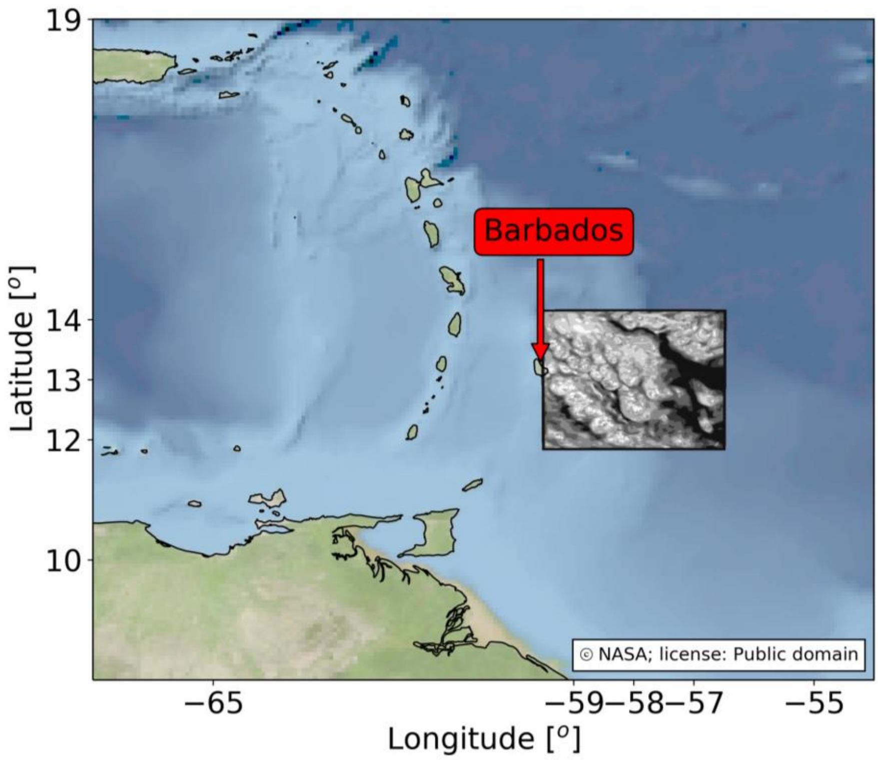

Figure 1. The domain of the simulations (the box in the middle) and the area around it. Inside the domain is presented the average cloud fraction over the first 30 mins of the simulation for 1/8/2016, CDNC = 20 cm^{-3}. The island of Barbados is marked with a red arrow.

Results

Conducting daily simulations over two months at different seasons allows us to sample a large ensemble of initial conditions and cloud types (see Fig. 2 and Table 1). To identify statistically significant differences between the two months, we conduct independent t-test (p-values are presented in Table 1). This demonstrates that the lower tropospheric stability (LTS), top of atmosphere shortwave flux (F_{SW}^{TOA}), and the atmospheric column radiative term (Q_R) are different in a statistically significant manner (p-value < 0.05) between the two different months. The differences in other parameters (cloud fraction – CF, liquid water path - LWP, ice water path – IWP, precipitation latent heat flux – LP, and top of atmosphere longwave flux - F_{LW}^{TOA}) are not statistically significant (Table 1).

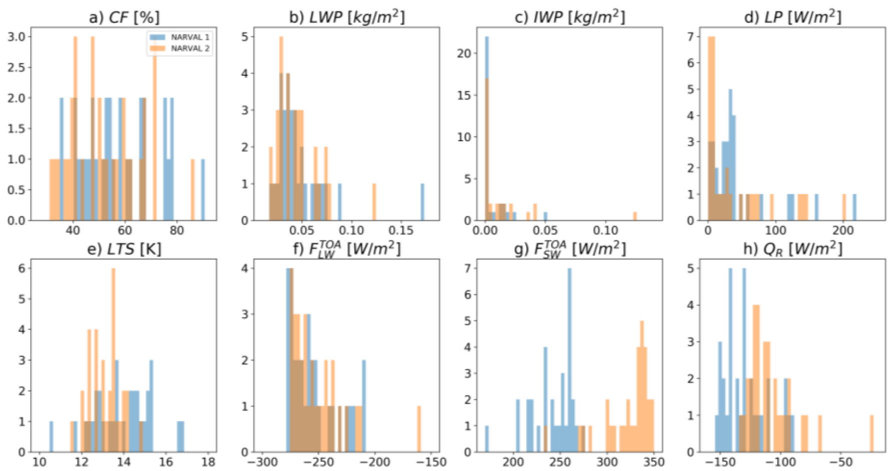

Figure 2. Histograms of mean (time and space) cloud and atmospheric properties for the base simulations with CDNC = 20 cm^{-3} (clean simulations) for each day of the two months that were simulated. Blue represents the NARVAL 1 month (December 2013), while orange the NARVAL 2 month (August 2016). a) cloud fraction – CF, b) liquid water path - LWP, c) ice water path – IWP, d) precipitation latent heat flux - LP, e) lower tropospheric stability – LTS, f) top of atmosphere longwave flux - F_{LW}^{TOA}, g) top of atmosphere shortwave flux - F_{SW}^{TOA}, and h) atmospheric column radiative term - Q_R.

Table 1. The monthly mean value of each of the properties presented in Fig. 2 ± 1 standard deviation for each month and the p-value of the two-sample independent t-test. The p-values which demonstrate a significant difference between the months (<0.05) are presented in bold.

	Mean NARVAL 1	**Mean NARVAL 2**	**p-value t-test**
CF [%]	57.2 ± 13.7	52.3 ± 13.4	0.16
LWP [kg/m^2]	$4.8 \cdot 10^{-2} \pm 2.8 \cdot 10^{-2}$	$4.5 \cdot 10^{-2} \pm 2.2 \cdot 10^{-2}$	0.66
IWP [kg/m^2]	$5.7 \cdot 10^{-3} \pm 1.1 \cdot 10^{-2}$	$1.2 \cdot 10^{-2} \pm 2.4 \cdot 10^{-2}$	0.19
LP [W/m^2]	43.8 ± 47.8	52.2 ± 78.2	0.6
LTS [K]	13.9 ± 1.4	13.1 ± 0.7	**$7 \cdot 10^{-3}$**
F_{LW}^{TOA} [W/m^2]	-254.2 ± 21.2	-251.7 ± 23.5	0.66
F_{SW}^{TOA} [W/m^2]	241.7 ± 22.5	321.9 ± 26.4	**$1.4 \cdot 10^{-18}$**
Q_R [W/m^2]	-129.2 ± 17.8	-107.8 ± 21.7	**$9.8 \cdot 10^{-5}$**

Figures 3 and 4 present vertical profiles of the total water (liquid and ice) mixing ratio from the different simulations during NARVAL 2 (August 2016) and NARVAL 1 (December 2013), respectively. Generally, during the winter month (NARVAL 1) the clouds are shallower than in the summer month (NARVAL 2), although there is significant variability. This is expected due to the seasonality of the ITCZ location (Stevens et al., 2016). The simulated days are manually separated to three different cloud regimes based on the domain and time mean total water mixing ratio vertical profiles. The cloud regimes considered here are: shallow clouds (shallow-cloud dominated days), two-layer clouds (shallow cloud layer and a cirrus cloud layer) and deep clouds (deep-cloud dominated days).

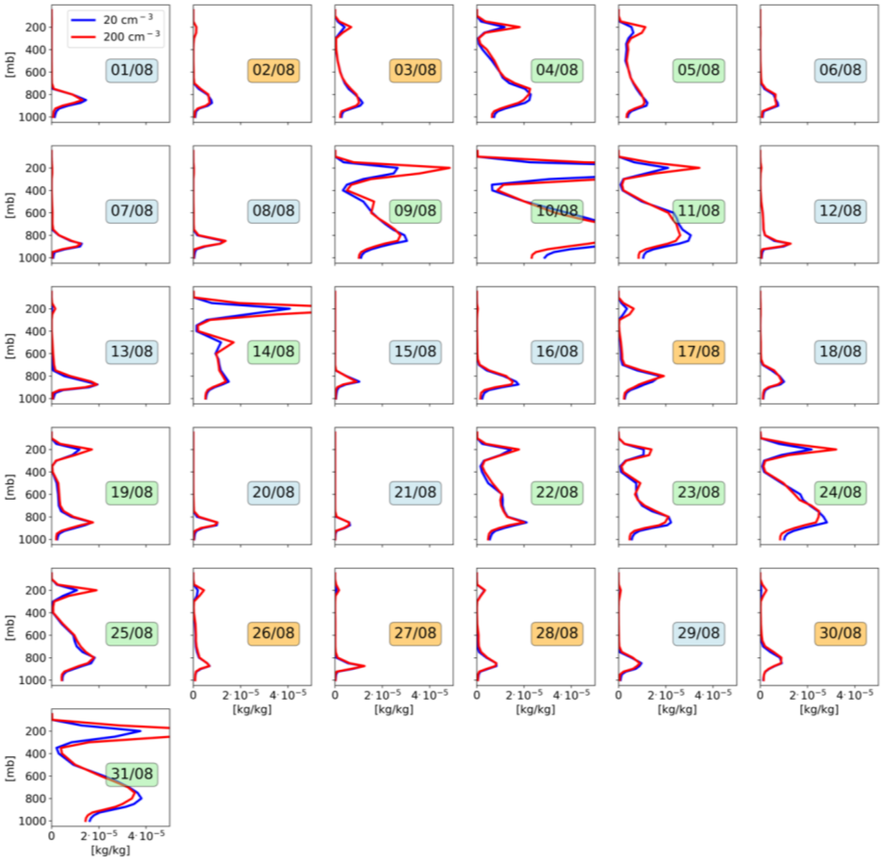

Figure 3. Mean (time and space) vertical profiles of the total water (liquid and ice) mixing ratio in each simulation (each last for 24 hours) for the NARVAL 2 month (August 2016). Blue: clean conditions (20 cm^{-3}), red: polluted conditions (200 cm^{-3}). The simulated days are separated into three different cloud regimes: shallow clouds (blue date box), two-layer clouds (shallow cloud layer and a cirrus cloud layer – orange date box) and deep clouds (green date box).

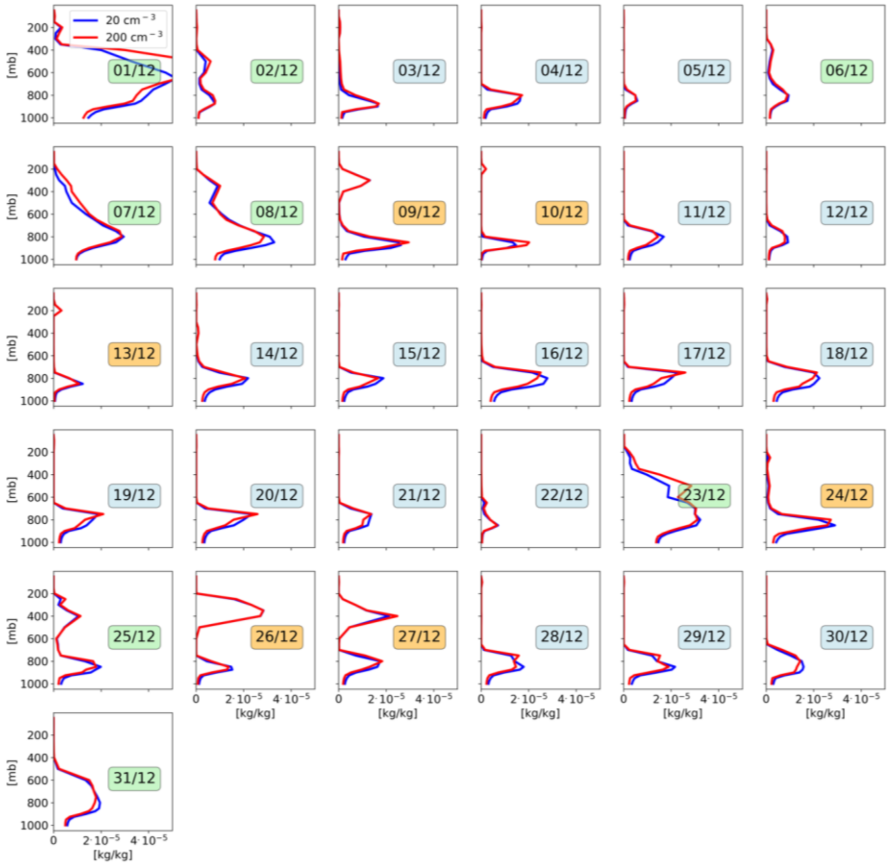

Figure 4. Same as Fig. 3 but for the NARVAL 1 month (December 2013).

Figure 5 presents histograms of aerosol effects (polluted minus clean) for the different simulations. The distribution of changes in cloud fraction (Fig. 5a) demonstrate small mean values for both months (-0.3% and 0.1% for the winter and summer month, respectively) which is slightly more skewed to positive values in the summer. Examining the significance of these trends with a t-test demonstrates that only the winter month response is statistically significant (Table 2). The CDNC effect on the liquid water path (LWP; Fig. 5b) and the ice water path (IWP; Fig. 5c) is shown to be almost entirely positive (or zero) in both months and differs from zero in a statistically significant manner. The mean change in precipitation (Fig. 5d) is small and negative (slightly more negative during the winter month). However, during the summer month it is not statistically significant and can be either positive or negative. We note that the mean precipitation decreases during the winter month (which is statistically significant) is small and equivalent to 0.07 mm/day (-1.8 W/m^2). Increasing CDNC systematically decreases LTS (Fig. 5e), representing deepening of the boundary layer (Dagan et al., 2016;Lebo and Morrison, 2014;Seifert et al., 2015;Stevens and Feingold, 2009). This trend is statistically significant for both months (Table 2).

The CDNC effect on $\boldsymbol{F_{LW}^{TOA}}$ is positive and small (average of 0.24 W/m^2) in the winter month (but still statistically significant) and larger (average of 2.16 W/m^2) in the summer month (Fig. 5f – positive flux downwards), primarily due to an increase in ice water content under polluted conditions (see also Figs. 3, 4 and 5c). We previously showed that an increase in CDNC drives an increase in the ice content at the upper troposphere and hence a reduction in the outgoing LW radiation (Dagan et al., 2019); here we show that this trend is statistically significant (Fig. 5c). However, during the winter, when deep convective clouds are less abundant and the atmosphere is more stable, the LW flux is less affected.

The CDNC effect on $\boldsymbol{F_{SW}^{TOA}}$ is always negative (Fig. 5g) and is on average -3.6 W/m^2 and -3.8 W/m^2 in the winter and summer month, respectively (the difference between the two months is not statistically significant; however, both differ from zero in a statistically significant manner -Table 2). The negative $\boldsymbol{F_{SW}^{TOA}}$ effect is caused mostly due to the Twomey effect (Twomey, 1977) and the LWP/IWP effect (Albrecht, 1989;Koren et al., 2010;Malavelle et al., 2017) (Figs. 5b and 5c), as the CF changes are small (Fig. 5a). For exploring the relative role of the Twomey and IWP/LWP effects, we ran all simulations again with the Twomey effect turned off. Without the Twomey effect the SW effect is reduced by up to a factor of 10 (-0.35 W/m^2 compared with -3.6 W/m^2 in the winter month, and -1.0 W/m^2 compared with -3.8 W/m^2 in the summer month). This demonstrates that the Twomey effect is the dominant factor underlying the $\boldsymbol{F_{SW}^{TOA}}$ changes. Radiative effects due to changes in ice size distribution are not considered due to uncertainties in the evolution of ice morphology. Accounting for this effect would likely further increase the relative role of the Twomey effect compare to the cloud adjustment effects (CF and LWP/IWP adjustments).

The change in the atmospheric column radiative warming term Q_R is shown to be small for the winter month (-0.26 W/m^2 on average) but much larger and positive for the summer month (1.8 W/m^2 on average). The increase in Q_R during the summer is caused due to the effect of deep, ice containing clouds on the outgoing LW flux (Fig. 5f). SW flux changes due to CDNC perturbations (Fig. 5g) have a much smaller effect on Q_R as the SW absorption of clouds is small (Dagan et al., 2019).

Examining the similarity between the response of the different properties to the CDNC perturbation in the two different months (Table 2) reveals that the responses of the IWP, $\boldsymbol{F_{LW}^{TOA}}$, Q_R and $\boldsymbol{F_{SW+LW}^{TOA}}$ (the net TOA LW and SW effects – Fig. 10 below) are different in a statistically significant manner between the two months. As will be shown below, this is related to the response of the ice content.

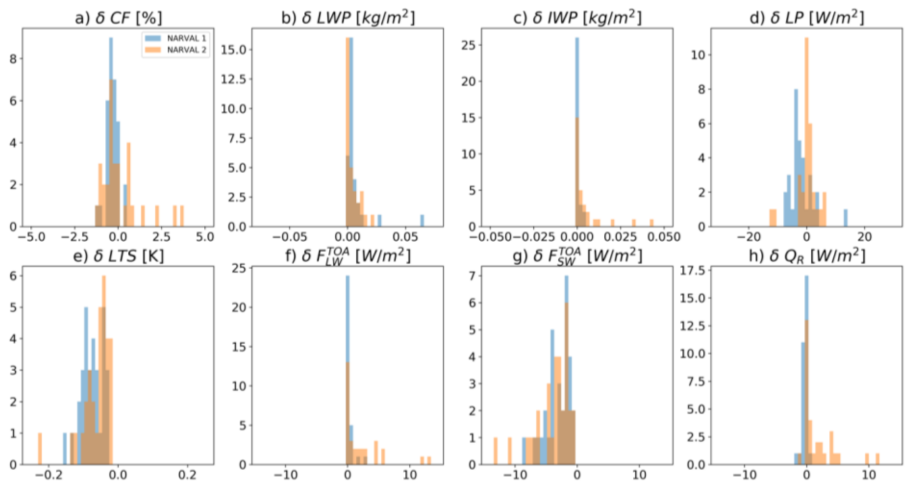

Figure 5. Histograms of the domain and time mean response of cloud and atmospheric properties to CDNC perturbation (polluted simulations minus clean simulations) for each day of the two months that were simulated. Blue represents the NARVAL 1 month (December 2013), while orange the NARVAL 2 month (August 2016). a) cloud fraction – CF, b) liquid water path - LWP, c) ice water path – IWP, d) precipitation latent heat flux - LP, e) lower tropospheric stability – LTS, f) top of atmosphere longwave flux - $\boldsymbol{F_{LW}^{TOA}}$, g) top of atmosphere shortwave flux - $\boldsymbol{F_{SW}^{TOA}}$, and h) atmospheric column radiative term - $\boldsymbol{Q_R}$.

Table 2. Summary of monthly mean response of cloud and atmospheric properties (presented in Fig. 5) to the CDNC perturbation (polluted simulations minus clean simulations) ± 1 standard deviation for each month. In addition, the p-values of the two-sample independent t-test are presented, as well as the p-values for comparing the CDNC response in each month to zero. The p-values which demonstrate significant difference (<0.05) are presented in bold.

	Mean NARVAL 1	Mean NARVAL 2	p-value t-test	p-value one sample t-test compare to 0 - NARVAL 1	p-value one sample t-test compare to 0 - NARVAL 2
δCF [%]	-0.32 ± 0.31	0.11 ± 1.15	0.053	**$8.1 \cdot 10^{-6}$**	0.6
δLWP [kg/m²]	$6.5 \cdot 10^{-3} \pm 1.2 \cdot 10^{-2}$	$4.0 \cdot 10^{-3} \pm 5.4 \cdot 10^{-3}$	0.3	**$4.4 \cdot 10^{-3}$**	**$3.5 \cdot 10^{-4}$**
δIWP [kg/m²]	$5.6 \cdot 10^{-4} \pm 1.3 \cdot 10^{-3}$	$8.2 \cdot 10^{-3} \pm 1.9 \cdot 10^{-2}$	**0.035**	**0.02**	**0.03**
δLP [W/m²]	-1.8 ± 4.1	-1.2 ± 7.0	0.7	**0.02**	0.37
δLTS [K]	-0.075 ± 0.031	-0.062 ± 0.042	0.18	**$3.2 \cdot 10^{-14}$**	**$4.3 \cdot 10^{-9}$**
δF_{LW}^{TOA} [W/m²]	0.24 ± 0.60	2.16 ± 3.25	**0.002**	**0.03**	**0.001**
δF_{SW}^{TOA} [W/m²]	-3.6 ± 3.5	-3.8 ± 2.9	0.8	**$3.3 \cdot 10^{-6}$**	**$4.7 \cdot 10^{-8}$**
δQ_R [W/m²]	-0.26 ± 0.39	1.8 ± 2.8	**$1.8 \cdot 10^{-4}$**	**$9.7 \cdot 10^{-4}$**	**$1.4 \cdot 10^{-3}$**
δF_{SW+LW}^{TOA}	-3.36 ± 3.02	-1.67 ± 1.93	**0.01**	**$1.1 \cdot 10^{-6}$**	**$5.1 \cdot 10^{-5}$**

CDNC effect on different cloud regimes

For better understanding the trend demonstrated in Fig. 5 and Table 2, we split the simulated days into different dominant cloud types/regimes (see Figs. 3 and 4). Figures 6 and 7 present histograms of the same atmospheric properties presented in Fig. 2 but separated by different cloud regimes – shallow clouds, two-layer clouds (shallow clouds with cirrus cloud layer above), and deep clouds. These figures demonstrate that the cloud fraction, LWP, IWP, precipitation, F_{LW}^{TOA} and Q_R are generally higher on days dominated by deep-clouds as compared to days dominated by shallow clouds, while the LTS and F_{SW}^{TOA} are lower in the deep-cloud dominated days compared to shallow-cloud dominated days (with the two-layer cloud days generally in-between them). The separation into different cloud regimes also demonstrates that more deep-cloud days are occurring during the summer month as compared to the winter month (12 compare to 8) and that the deep clouds during summer are deeper and contain more water. The larger occurrence of deep convection during the summer month is consistent with the statistically significant reduction in LTS (Fig. 2 and Table 1) and is expected based on the local seasonality (Stevens et al., 2016).

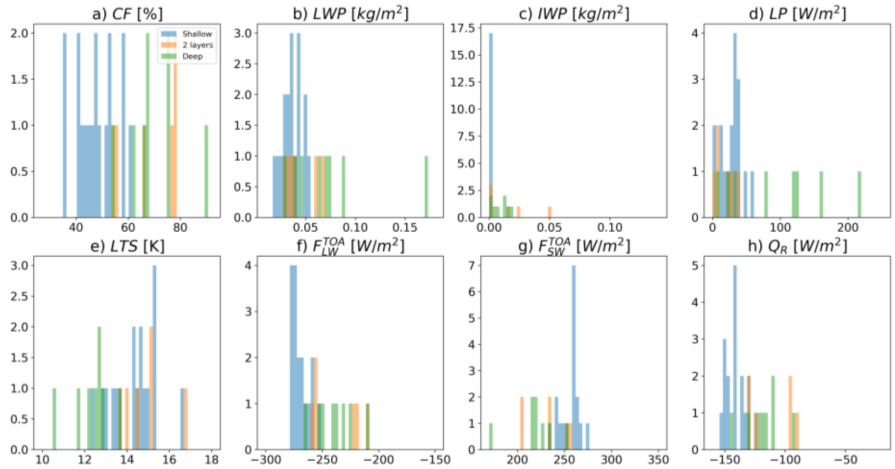

Figure 6. Histograms of mean (time and space) cloud and atmospheric properties for the base simulations with CDNC = 20 cm^{-3} (clean simulations) for each day of the NARVAL 1 month (December 2013) separated into different cloud regimes: shallow clouds (blue), two-layer clouds (shallow clouds with cirrus clouds layer above - orange), and deep clouds (green). a) cloud fraction – CF, b) liquid water path - LWP, c) ice water path – IWP, d) precipitation latent heat flux - LP, e) lower tropospheric stability – LTS, f) top of atmosphere longwave flux - F_{LW}^{TOA} , g) top of atmosphere shortwave flux - F_{SW}^{TOA}, and h) atmospheric column radiative term - Q_R.

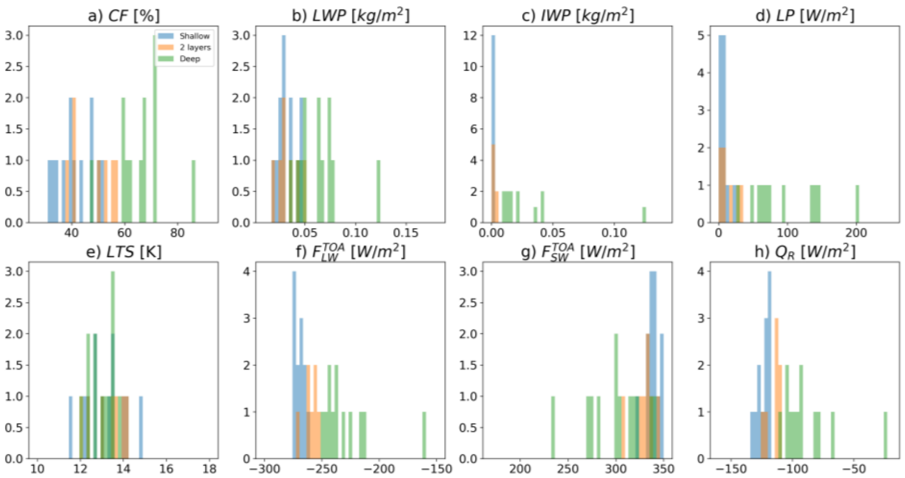

Figure 7. Same as Fig. 6 but for the NARVAL 2 month (August 2016).

Examining the response of the different cloud regimes to the CDNC perturbation (Figs. 8 and 9) demonstrates that the response of the cloud fraction, LWP, IWP and F_{LW}^{TOA} in the deep-cloud days is generally more positive, while the response of F_{LW}^{TOA} and LTS is generally more negative. These trends are more pronounced during the summer month as compared to the winter month. The response of Q_R is more positive in the deep-cloud dominated days in the summer month but does not show any different trend in the winter month. The precipitation response does not show any distinct different trend for the different cloud types in both months.

The findings presented in Figs. 8 and 9 demonstrate that the IWP response in the deep-cloud dominated days is generally stronger in the summer month as compare to the winter month. The increase in the IWP with the increase in CDNC drives a reduction in F_{LW}^{TOA} and hence increase in Q_R (Dagan et al., 2019). We note that the largest difference between the two months emerges due to the stronger response of the ice content in the summer month as compared to the winter month. This fact can explain the statistically significant different response of the IWP, F_{LW}^{TOA} and Q_R shown in Table 2.

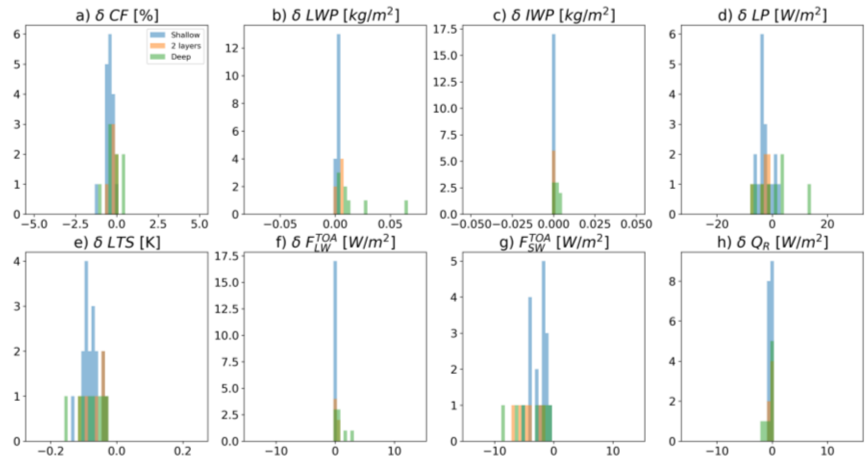

Figure 8. Histograms of the domain and time mean response of cloud and atmospheric properties to the CDNC perturbation (polluted simulations minus clean simulations) for each day of the NARVAL 1 month (December 2013) separated into the different cloud regimes: shallow clouds (blue), two-layer clouds (shallow clouds with cirrus clouds layer above - orange), and deep clouds (green). a) cloud fraction – CF, b) liquid water path - LWP, c) ice water path – IWP, d) precipitation latent heat flux - LP, e) lower tropospheric stability – LTS, f) top of atmosphere longwave flux - F_{LW}^{TOA}, g) top of atmosphere shortwave flux - F_{SW}^{TOA}, and h) atmospheric column radiative term - Q_R.

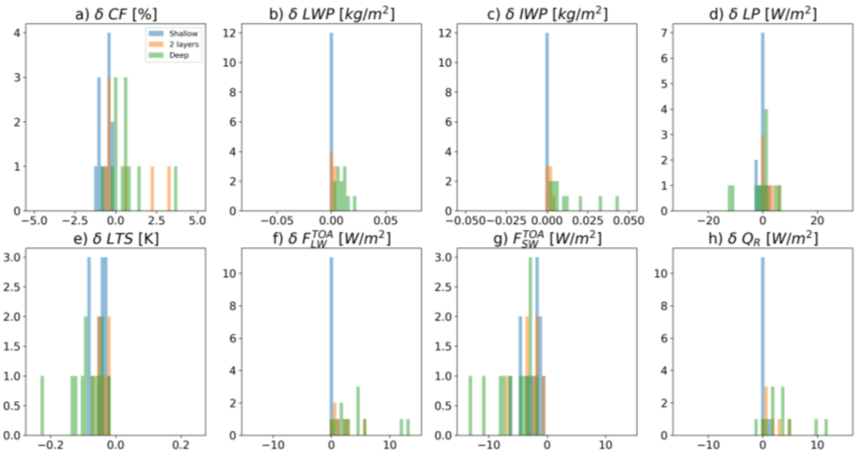

Figure 9. Same as Fig. 8 but for the NARVAL 2 month (August 2016).

The combined CDNC effect on the total net TOA radiation (F^{TOA}_{SW+LW}) is shown in Fig. 10. It demonstrates that during the winter month the effect on F^{TOA}_{SW+LW} is always negative and has a mean value of -3.4 W/m^2. However, during the summer month, the mean effect is less negative (-1.7 W/m^2) and for some of the days it could even be positive due to the effect of the CDNC on the ice water content (Fig. 5 and Table 2). The difference between the two months in F^{TOA}_{SW+LW} is statistically significant (Table 2). We note that during the summer month all days for which $F^{TOA}_{SW+LW} \geq 0$ are deep-cloud dominated days, supporting the hypothesis that the difference between the different months are driven by the different response of the deep clouds, which are deeper and contain more water in the summer month.

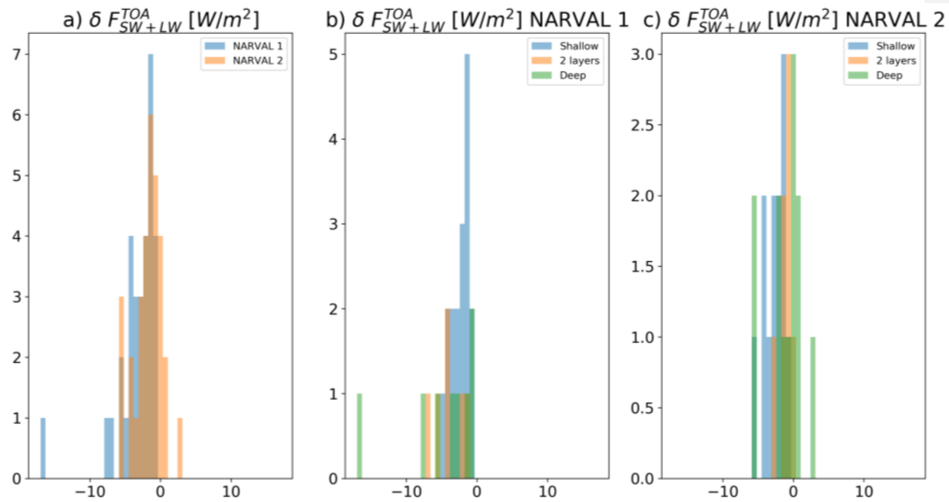

Figure 10. Histograms of the response of the net (shortwave + longwave) top of atmosphere radiative flux (F^{TOA}_{SW+LW}) to the CDNC perturbation (polluted simulations minus clean simulations) for each of the simulated days. In a) blue represents the NARVAL 1 month (December 2013), while orange the NARVAL 2 month (August 2016). In b) and c) the NARVAL 1 and the NARVAL 2 months are separated to the different cloud regimes: shallow clouds (blue), two-layer clouds (shallow clouds with cirrus clouds layer above - orange), and deep clouds (green).

Summary

Ensemble daily simulations over a region near Barbados for two separate month-long periods were conducted to investigate aerosol effects on cloud properties and the atmospheric energy budget. For each day, two simulations were conducted with low and high CDNC representing clean and polluted conditions, respectively. These simulations are used to distinguish between properties that are robustly affected by changes in CDNC and those that are not. For example, we have shown that, for the entire set of simulations (62 different days), an increase in CDNC always drives a reduction in the lower tropospheric stability (Fig. 5). In addition, F^{TOA}_{SW} is always reduced by an increase in CDNC, representing more SW reflection. However, changes in cloud fraction or precipitation are not as robustly affected, and, despite the fact that for a given day they could be large, on average they are not distinguishable from zero (at least for the summer month). However, we note that the aerosol response we present here may be underestimate due to the effect of the fixed boundary conditions~~ and hence is estimated as the lower bound~~. In addition, using a microphysical scheme that assumes saturation adjustment reduces the sensitivity of the clouds to aerosol perturbation (Koren et al., 2014; Dagan et al., 2015a;

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Heiblum et al., 2016; Fan et al., 2018). We also note that using 1200 m horizontal resolution does not properly resolve all shallow cumulus clouds.

TIn addition, the use of two month-long periods, covering different seasons dominated by different meteorological conditions and cloud type, demonstrate again (Altaratz et al., 2014;Lee et al., 2009;Mülmenstädt and Feingold, 2018;van den Heever et al., 2011;Rosenfeld et al., 2013;Glassmeier and Lohmann, 2016;Gryspeerdt and Stier, 2012;Dagan et al., 2015a), that the aerosol effect on clouds is strongly dependent on cloud regimes and meteorological conditions. For our simulations we demonstrate that the top of atmosphere net radiative effect is twice as large during the winter month as compared to the summer month (Fig. 10).

To better understand these differences we have split the simulated days into three different dominant cloud regimes. The results demonstrate that most of the differences in the response to CDNC increases between the two months are driven by the response of the ice content in deep convective clouds. During the summer month, the atmosphere is less stable and the deep convective clouds in the base-line simulations are more abundant, reach higher levels in the atmosphere and contain more water. These more developed clouds respond stronger to the CDNC perturbations and develop more ice content than the shallower clouds during the winter month. The increased ice is driven by increase in mass flux to the upper levels. The added ice content reduces the outgoing LW flux at the TOA and hence compensates some of the SW effect, which itself is similar between the summer and winter months.

Our results highlight the need to use large ensembles of initial conditions for cloud-aerosol interaction studies, even in large domain simulations, and suggest that caution is needed when trying to draw conclusions from a single case-study experiments and short-term observations.

Author contributions. G. D. carried out the simulations and analyses presented. P. S. assisted with the design and interpretation of the analyses. G.D. prepared the manuscript with contributions from P.S.

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