We thank both reviewers for their comments. We address each in turn below, with responses indented, and revised text in red.

Reviewer 1

I think this is a significantly improved manuscript. It's very close to publication quality. I think this manuscript deserves to be highlighted, and I thus I reviewed it below as a highlights candidate. In my opinion, it requires a bit of editing to make it highlights worthy, which I hope the authors agree with. I list my comments below for potential improvements, and I hope the authors find them constructive. All my comments below should be considered minor (unless the authors deem them otherwise) and they should be considered non-blocking for publication.

> We are delighted by this summary and very pleased that our revisions have addressed the reviewer's concerns and helped to improve the manuscript. We agree with all of the suggestions that the reviewer has provided and have made appropriate revisions. We hope that the revised manuscript has been enhanced to the level required to be considered a highlights candidate, as suggested by the reviewer.

Minor comments:

The manuscript is still hard to read; if the authors can spare some time improving readability and presentation, that will go a long way. In general, it's improved significantly, and it is publishable as-is, but it could be improved and should ideally be highlighted (because it is a pretty good study)

> We have made changes throughout the manuscript to improve the readability. This includes revisions in response to the reviewer comments and additional revisions.

There several minor/technical issues that may warrant minor fixes (I list all of these below). I encourage the authors to fix them

I think the authors downplay the usefulness of their idealized setup, MACv2-SP. I think it would be nice if the authors make it clear where they see advantages in simplified schemes beyond computational performance (e.g., causality, process isolation) and where they see shortcomings (e.g., non-interactive nature of aerosols and lack of mesoscale features shown in ICON-HAM-Lite). The authors do plenty of the latter (shortcomings) but not enough of the former (advantages). After all, you're using a simplified scheme and if the only reason you're doing this study is because you had to, then that's no good. I think there's value in these simplified schemes in that they allow us easier access to some process aspects that are significantly harder to disentangle with fully interactive schemes. If you disagree with me, feel free to ignore. I just feel it is a missed opportunity not to forcefully defend these simplified schemes as practical and appropriate for some endeavors (you could also make your argument by carefully citing Stevens et al and Fiedler et al papers where they described the original MACv2-SP scheme)

We agree with the reviewer and have included a new paragraph in the introduction that outlines the benefits of using an idealized representation of aerosols. The new paragraph is below.

“Aerosols themselves are also a source of uncertainty in ESMs and high-resolution simulations due to complex aerosol microphysical processes that are poorly constrained or inadequately represented (White et al., 2017; Sand et al., 2021; Vogel et al., 2022; Regayre et al., 2018; Gliß et al., 2021). This complexity can also inhibit the interpretability of model behavior (Proske et al., 2023) and may not necessarily scale with improved model representation (Ekman, 2014). Previous studies have used idealized or simplified aerosol representations to remove this uncertainty and focus on quantifying aerosol interactions at the process level. Prescribed aerosol fields have been used to systematically quantify the sensitivity of the atmosphere to aerosol properties, including horizontal gradients (Lee et al., 2014), vertical profiles (Herbert et al., 2020; Johnson et al., 2004), concentrations (Dagan and Eytan, 2024; Tang et al., 2024), and spatial distributions (Williams et al., 2022; Dagan et al., 2021; Fiedler et al., 2017; Herbert et al., 2021a; Fiedler and Putrasahan, 2021). Idealized aerosol representations have also proven useful for identifying model structural uncertainties and estimating aerosol radiative forcing in intercomparison studies (Stier et al., 2013; Fiedler et al., 2019; Randles et al., 2013; Fiedler et al., 2023) and have been combined with reduced complexity climate models to provide a means of assessing sensitivity to future aerosol scenarios (Herbert et al., 2021b; Stjern et al., 2024; Recchia and Lucarini, 2023).”

We have also revised a paragraph in the conclusions to positively frame our choice:

“The idealized representation of aerosol in this model has helped identify important process-level interactions and provides a platform for future studies using realistic aerosol perturbations. The use of non-interactive aerosol may mask important feedbacks…”

Finally, I think the discussion around convection could be improved, but I will admit (like I do below) that this is not my area of expertise, so I cannot judge the claims sufficiently. After reading these parts a few times, I felt the arguments were wishy washy and not very convincing when it comes to convection. They strongly imply significant convection changes at times, but other times the authors caution over-interpretation. I would encourage a careful reread of those parts dealing with convection, then assessing if more careful phrasing and revision could be used. I also suggested referring (and/or reminding) the readers to a convection assessment of ICON (if at all). Would any conclusions about aerosol–convection interactions be affected if the convection itself isn't as good/robust?

The manuscript already includes references that demonstrate the ability of our ICON configuration to represent convection. In the revised manuscript we have expanded this and refer to the relevant section later in the manuscript. See the response to comment further on.

The suppression of convection via changes to the convective environment are a consistent result throughout the analysis and are in agreement with previous studies. We only apply caution to changes in convection that are related to cloud microphysical processes (e.g., convective invigoration), which our results suggest is not consistently occurring in all regions. The role of convective invigoration remains the focus of many research groups.

To avoid confusion, we have revised the manuscript to clearly separate the responses to the convective environment from the direct modification to convection occurring within the clouds.

This includes the following revisions in Section 3.2.2:

"Some modelling studies have suggested aerosols can also directly influence convection through invigoration of convective cloud cores via ACI, either in the liquid phase (Lebo, 2018; Sheffield et al., 2015; Fan et al., 2018) or ice phase (Heever et al., 2006; Fan et al., 2013), whilst others report suppression or regime-dependence Khain et al. (2008); Lebo and Seinfeld (2011); Storer et al. (2010); Igel and van den Heever (2021)."

"The latter is consistent with the impacts to the convective environment as observed over the Amazon and the Congo, while the former is a modification to the large-scale circulation."

"This occurs alongside an increase in IWP and θ, which suggests a role for direct modification of the convective cloud cores via convective invigoration from the cold phase (Heever et al., 2006; Fan et al., 2013)."

And the following the conclusions section:

"Three regions, characterized by deep convection and emissions of biomass burning aerosol, consistently demonstrated a suppression of the diurnal cycle of convection via modifications to the convective environment due to ARI and enhanced LWP due to ACI. However, the combined effect (ARI + ACI) differed in each region. The direct modification to convective clouds (suppression or invigoration) via ACI also differed between regions."

More comments:

L9: It's not readily clear to me how this sentence implies anything about atmospheric dynamics — even with controlled dynamics (say nudged simulations), we will see strong regional dependence, no? I'd recommend removing it from the abstract unless you can justify it later? I didn't really see enough strong evidence supporting it. I would remove the part about atmospheric dynamics especially that the next point is by far the least controversial and most important point of this manuscript. Note my point here isn't debating if the statement is true in general (I think it is true; e.g., I agree with your framing near L275), but rather, your manuscript/results don't have enough evidence to support "complex interplay with atmospheric dynamics" highlight imo

Line in question: "*In our simulations over 30 days, we find that the aerosol impacts on clouds and precipitation exhibit strong regional dependence, **highlighting the complex interplay with atmospheric dynamics**"*

We agree with the reviewer. Our results suggest that most of the regional dependence seems to be driven by the large-scale environmental factors, which shape the underlying properties of the region. The reviewer refers to L275 which may be: *This demonstrates that the impact of aerosol on clouds has a diurnal driver, that may be dependent on the underlying diurnal cycle of clouds, dynamics, or solar radiation.*

As suggested, we have removed the statement.

L10: Imho, this is the most important point of this manuscript. I think this alone justifies the manuscript and effort. It is also highlighted in the title. Great work!

The reviewer refers to the sentence "The impact of ARI and ACI on clouds in isolation shows some consistent behaviour, but the magnitude and additive nature of the effects are regionally dependent."

We thank the reviewer for this comment.

L13: This may benefit from a slight clarification to drive the point home stronger. I think you're trying to say something like "Because we observe pronounced diurnal cycles … we think polar-orbiting satellites may be even more limited than we already know." If so, I would rephrase to something like ", suggesting the usefulness of using polar-orbiting satellites to quantify ACI may be even more limited than presently assumed"

Lines in question "*We also observe pronounced diurnal cycles in the response of cloud microphysical and radiative properties, suggesting a limitation of using polar-orbiting satellites to quantify or constrain aerosol-climate interactions on the diurnal scale"*.

We have rewritten the sentence as suggested:

"We also observe pronounced diurnal cycles in the response of cloud microphysical and radiative properties, which suggests the usefulness of using polar-orbiting satellites to quantify ACI and ARI may be more limited than presently assumed."

L17: I found the statement about ACI/ARI in the Conclusion section (e.g., L485) to be quite important and remarkable and I thus recommend including more about it in the abstract (maybe as a follow-up sentence to the great sentence on L10?)

From the conclusions section: "*The results also strongly suggest that ACI and ARI cannot be considered independently as the responses via each pathway does not tend to be additive. Some were dominated by either ACI or ARI, and some behaved non-linearly, resulting in a response at odds with the individual components*."

We have expanded the abstract sentence in question. The revised text is as follows:

"The impact of ARI and ACI on clouds in isolation shows some consistent behavior, but the magnitude and additive nature of the effects are regionally dependent. Some regions are dominated by either ACI or ARI, whereas others behaved nonlinearly. This suggests that the findings of isolated case studies from regional simulations may not be globally representative and that ARI and ACI cannot be considered independently and should both be interactively represented in modelling studies."

L86: Usually, models prescribe the sea ice extent, but let the sea-ice thermodynamics run. If that's the case in your model, I would simply add "extent" or "cover" after sea ice to avoid confusion

The line in question is "We run the model in an atmosphere-only mode, with oceanic properties (sea surface temperature and sea ice) prescribed following the atmospheric model intercomparison project AMIP"

The model does not have a sea-ice thermodynamic model and instead uses the prescribed sea-surface properties as boundary conditions, in-line with the AMIP protocol. We have rephrased this sentence as follows:

"*We run the model in an atmosphere-only mode, with sea surface properties (sea surface temperature and sea ice **concentration**) prescribed **as atmospheric boundary conditions** following the atmospheric model intercomparison project AMIP"*

L117: I would mention the exact process (which you do later anyway, but might as well do it here too: autoconversion)

Line in question: "*In this study, aerosols are represented using the simple plume implementation of theMax Planck Institute Aerosol Climatology version 2 (Stevens et al., 2017, MACv2-SP), which is used in ICON to represent aerosols in the radiation scheme. We extend this to the cloud microphysics scheme to link changes in aerosol to the warm-rain process"*.

This has been included as suggested.

L118: I thought they are non-interactive but spatially and temporally varying? Like AOD is different over Africa compared to Poles. Same for Nc and/or Nd. Are you talking about something else here? When I read "fields … provided by MACv2-SP" I think of fields that MACv2-SP prescribes for your simulations (optical depths + Nd + Nc). Maybe you're talking about the inputs to the plume model or something else here…

Line in question: "*The prescribed fields of aerosol provided by MACv2-SP are non-interactive and spatially invariable, but magnitudes are temporally variable."*

The reviewer is correct: the aerosol fields provided by MACv2-SP are spatially and temporally variable. The phrasing above was aimed to clarify that once prescribed, the spatial distribution is set. This is misleading, so has been rewritten for clarity:

"The prescribed fields of aerosol provided by MACv2-SP are non-interactive, but magnitudes are spatially and temporally variable."

L164: Minor point: is it more or less consistent with obs than the prior tuning?

This refers to the parameters a_N and b_N which are used to calculate $N_{d,cld}$ from the AOD. In response to the second reviewer, we expanded the N_d climatology in Figure 1b. In the

Grosvenor et al. (2018) manuscript, N_d was shown over oceans only, but the authors also provide data over land in the supporting information. This provides a better test for our aerosol representation as the MACv2-SP plumes are centered over land (where primary aerosol emissions occur), not the ocean. We used this alternative dataset and removed grid boxes with relatively large uncertainty - where the number of days with a successful retrieval over the 13 year time series was less than 50.

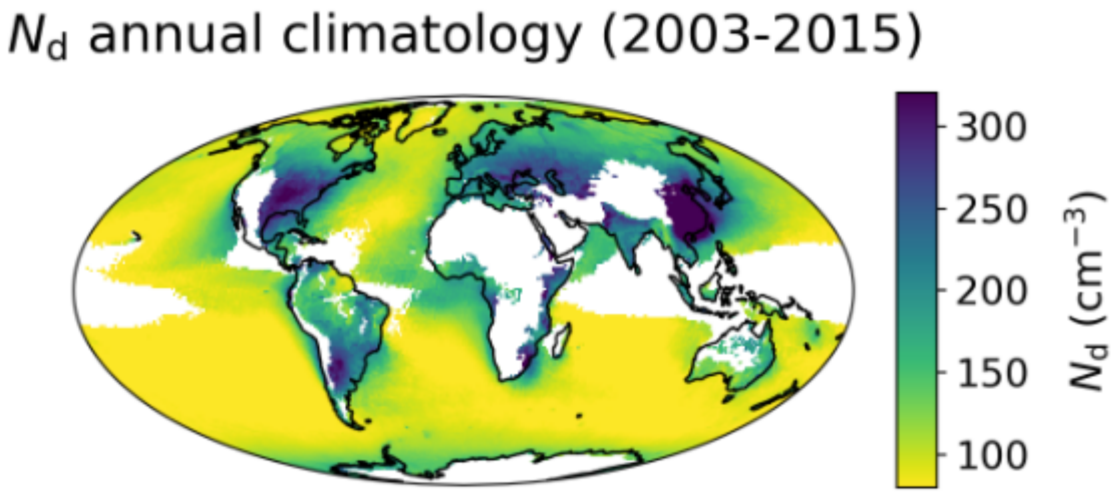

Figure R1. Revised annual mean N_d climatology from Grosvenor et al. (2018). Blank regions show grid boxes where there were fewer than 50 retrievals in the 13 year time series.

A statistical comparison to the climatology using the new and default parameters is shown in Table R1 below. All statistical tests demonstrate that the new parameters are more appropriate than the default ones. Using the more expansive N_d climatology has also increased the correlation coefficient from 0.34 (oceans only - unconstrained) to 0.57 (all available grid points - constrained to > 50 data points). This value still demonstrates some discrepancy between simulation and observations but provides more confidence to the reader.

	Herbert et al. (2021) parameters	Default (Stevens et al., 2017) parameters
Root mean square error	49.5	70.6
Mean absolute error	34.0	47.3
Normalized mean bias	-0.12	-0.35
Correlation coefficient	0.57	0.42

Table R1. Statistical tests between observed N_d from Grosvenor et al. (2018) and simulated N_d using the MACv2-SP plume model.

The revised climatology has been included in the revised manuscript, and the following text has been included in Sect. 2.3:

"A comparison between simulated and observed N_d yields a root mean square error (RMSE) of 49 cm^{-3} and a correlation coefficient of 0.57 (the default parameters a_N and b_N yield an RMSE of 70 cm^{-3} and correlation coefficient of 0.42). The discrepancy is in part due to high simulated values over biomass burning regions that are not reflected in annual mean observations, but also due to regional variability that MACv2-SP does not capture (e.g., North America)."

Figure 1b has also been updated to Fig R1 above and the caption has been expanded to describe the new method:

"... and (b) annual mean N_d for cloud tops < 3.2 km (2003 – 2015; only showing grid points with 50 successful retrievals) estimated by Grosvenor et al. (2018)..."

L172: Minor: I would consider using the table to specify the details very clearly and anchor the discussion around it. For example, PI run uses Nd,cld that's constant (value xyz), but Nd,rad that evolves according to Eq X with constant values for xx and yy. Currently, you do have all the info in the text, it is just slightly hard to parse. But feel free to ignore…

This is an excellent suggestion. Table 1 has been expanded and provides more detail. We have also rewritten the opening paragraph of Section 2.3:

"We use four simulations to explore the role of aerosols on clouds and climate (outlined in Table 1). The control simulation (PI) uses values that are representative of a pre-industrial atmosphere consisting of natural aerosol and background ARI and ACI effects. Global fields of natural aerosol extinction are represented by the K19 climatology for the year 1850. $N_{d,cld}$ is held constant at a value of 80 cm^{-3}, whilst $N_{d,rad}$ follows a vertical profile according to Eq. 1 and varies spatially with $N_{d,rad-sfc}$ set to 120 cm^{-3} on land and 80 cm^{-3} over oceans. A second simulation (PD) is run with values that are representative of a present-day atmosphere that includes ACI and ARI effects due to anthropogenic activity. Aerosol extinction fields from anthropogenic aerosol are represented by the plume model MACv2-SP for the year 2016 and added to the pre-industrial contribution. The spatial distributions of $N_{d,cld}$ and $N_{d,rad}$ are modified using the scaling factor f_N (Eq. 2), which varies spatially with the anthropogenic aerosol contribution. The third and fourth simulations are used to isolate ACI and ARI effects in the present-day atmosphere. In the third simulation, PD_{ARI}, extinction from the anthropogenic aerosols are included, but the scaling factor f_N is not applied to $N_{d,cld}$ and $N_{d,rad}$; this isolates ARI effects associated with anthropogenic aerosol. In the final simulation, PD_{ACI}, the ACI scaling factor f_N is applied, but aerosol extinction remains at pre-industrial values; this isolates ACI effects associated with anthropogenic aerosol."

L200–215: Great! I think this is very promising and appropriate!!

This comment refers to the lines where we outline that we do not focus on global aerosol forcing estimates, and instead "*exploit the capability of the model to represent scales that are traditionally used by high-resolution simulations*".

This was the new focus of the study as suggested by all three reviewers. We are very happy that the reviewer positively acknowledges these revisions.

L301 (and many other places): You use the word "response" a lot in this manuscript (on one editor, I found 117 instances when I used the pdf search functionality) and you use it to mean at least two different things. On this line, you likely mean just the values of the CFtotal in Figure 8. But elsewhere you use it to mean the PD-PI response (other places you also use in the second sense, related to change). Can you try to clarify this throughout the manuscript? For example, in Figure 7 (just above) you likely just mean "… during diurnal cycle of maximum absolute for each..." And on this line you like just mean "spatially consistent with CFtotal, with" … unless I missed something obvious?! I bring this up because the word response tripped me up multiple times, and I got confused trying to understand what you're trying to say. I found myself having to go back and cross-check. My recommendation is to use the word "response" only in making an explicit point about "a response to a perturbation" (e.g., your L308, or caption of Figure 10). For everything else, I'd use different ways to describe the signal more matter-of-factly/plainly

We use the word response to refer to the change in the variable due to the aerosol perturbation (e.g. PD - PI). In the three examples used in the reviewer comment we are indeed referring to the response. In response to this comment, we have gone through the manuscript to break up the repetitive use of the word response. In some instances response has been changed to the 'aerosol effect' and in others the 'change in X' or 'ΔX'. We have also checked for instances where its use is either redundant or potentially misleading - for example, we often use the term "...in the PD response". These have been revised for clarity.

L319: Minor, but has there been an assessment of deep convection characteristics in this model? Is it okay compared to obs? I am not a convection expert and I don't study convection–aerosol interactions, but I keep hearing that 5km models have pretty severe convection problems, so I am not sure how informative it is to study the aerosol impacts on or due to convection, if the convection is poorly simulated to begin with. This is borderline outside your scope here, so my only minor request is to add (somewhere in the manuscript, if not added already) some references about a convection assessment of this specific model config if one exists…

Line in question: The novel aspect of this study is the globally resolved deep convection, hence we primarily focus on regions with deep convection.

Following comments in the first round of reviews, we expanded a paragraph in the introduction to discuss the model's ability to represent convection. The paragraph is as follows:

"This configuration of ICON does not explicitly resolve the smallest scales of convection (< 5 km) but has been shown to reproduce observed diurnal and seasonal cycles of tropical precipitation (Segura et al., 2022). Given that ESMs tend to use spatial resolutions of tens to hundreds of kilometres, this makes a marked improvement in our ability to resolve many aspects of convection (Done et al., 2004; Prein et al., 2013) and is well suited for our study."

We have slightly expanded this as follows and included a pointer to the section (Sect. 2.1) on the relevant line.

"*This configuration of ICON does not explicitly resolve the smallest scales of convection (< 5 km) but has been shown to reproduce* ***many features of the climate system relevant for this study (Hohenegger et al., 2023), including seasonal cycles of precipitation and soil moisture, the structure of the atmosphere in deep convective regions, and coupling between sea surface temperature and precipitation.*** *Segura et al., (2022) also demonstrate that this configuration reproduces the observed diurnal cycle of tropical precipitation. Given that ESMs tend to use spatial resolutions of tens to hundreds of kilometres, this makes a marked improvement in our ability to resolve many aspects of convection (Done et al., 2004; Prein et al., 2013) and is well suited for our study."*

L375 (and thereabouts): I don't think these variables (Mflux, W*) were defined/introduced, but I may have missed them… I would introduce them with equations or a reference to an equation/methodology elsewhere

M_{flux} is defined in Section 3.1 but W* is only defined in a figure caption. The latter has been moved to the main text along with an introduction to the other variables shown in Figures 11, 12, and 13:

"In Figures 11 – 13 we focus on the drivers of the cloud response to anthropogenic aerosol in the three convective regions. Variables include IWP, P, and M_{flux}, and profiles of ice water content (IWC), liquid water content (LWC), potential temperature (θ), water vapor (Qv) and vertical velocity (W) calculated in regions characterized by ascent (1° grid boxes where the mean vertical velocity at 300 hPa during the PI simulation is positive). The frequency of output on all vertical…"*

L397: which state? Here and elsewhere, I would try to be explicit about which state you mean (dynamic state, thermodynamic state, aerosol state, cloud state, or general atmospheric state, or something totally different?)

Line in question: "*The Congo and Amazon regions respond consistently to ARI and ACI individually, but the total responses are different, suggesting a degree of state dependence"*.

These instances have been clarified as *thermodynamic states* and *thermodynamic state dependence*.

L415: Related to the above: you start with localized modification to the convective environment, but here you say convection itself is modified. How did the jump take place? Is the reader supposed to assume that the localized modifications to the convective environment will always lead to convection changes?

L410: Like above, here and elsewhere, I would try to be explicit about what you mean by convective environment. Of course, you can define it the first time you mention it and say "convective environment" is my shorthand for what I just defined throughout the manuscript.

We have combined these two comments.

The *convective environment* refers to the properties of the local atmosphere that describe the potential for initiation and development of convection. This will include vertical profiles of temperature, moisture, and wind shear, and larger-scale horizontal fluxes (convergence/divergence). If this environment is made more or less favourable for the initiation and/or development of convection then it follows that there will be changes to convection. The aerosol perturbations can modify these properties through changes to surface fluxes and heating profiles. The aerosol may also directly modify convection via cloud microphysical processes that are linked to the availability of aerosol (CCN, cloud droplets, latent heat, buoyancy).

As suggested, we have defined the term *'convective environment*' upon first use:

"However, this will also be sensitive to the thermodynamic properties of the region that provide the potential for convection (the convective environment), the different aerosol plume characteristics, or buffering of the response due to coupling to large-scale meteorology."

We have also expanded/revised some sentences for clarity and to help distinguish the convective environment from convection itself:

"The strongly absorbing aerosol produces localized heating of the smoke layer, suppressing mixing in the lower atmosphere and drying aloft, which reduces the potential for convection in the region. The suppressed convection reduces the regional-mean vertical extent of clouds and decreases LWC throughout the column."

"...suggests the differences may be due to stronger capacity to buffer the perturbation over the Congo, which tends to exhibit more convection than the Amazon, or differences in the convective environments Storer et al. (2010) that may result in one region being more susceptible to the aerosol perturbation."

"The contrasting roles of ACI and ARI in the Congo and the Amazon suggest that the response of convection to changes in the aerosol population is dependent on the background thermodynamic state and convective environment, which has also been observed in remote-sensing studies of the Amazon region.."

"The primary driver is a localized modification to the convective environment that suppresses convection and reduces daily accumulated P…"

L498: I think I finally get what you're saying with the "spatially invariable" part… the spatially invariable aerosol as input to MACv2-SP? But this is quite misleading! Your model doesn't really care about that aerosol input to the MACv2-SP model; what your model cares about is the effect of the aerosol (as proxied by radiation properties like

optical depths, Nd,cld, and Nd,rad). These are definitely spatially variable! Am I misunderstanding something here?

> This comment was echoed previously. The aerosol fields provided by MACv2-SP are spatially and temporally variable. The phrasing of ‘spatially invariable’ was aimed to clarify that once prescribed, the spatial distribution is set. This is misleading, so has been rewritten for clarity.

L502: I would delete this last sentence advertising modeling groups (doesn’t really add any context beyond what you said)

> As suggested, the sentences have been removed.

L520: I would remove this entire paragraph (ending on the prior paragraph is much better imho).

> As suggested, the paragraph has been removed.

L527: Could you provide a specific commit/sha for the code you used? Or even better include it in a permanent Zenodo repository with its own DOI? Thanks!

> This information has been added to the data availability section.

Reviewer 2

The authors addressed some of my comments in the last round of review, but the manuscript still suffers from severe flaws despite being mostly rewritten.

Firstly, the authors used LOESS, a tool for seasonal-trend decomposition, to separate the long-term component from the short-term component in the model time series. They insisted that the long-term component comprised internal variability and persistent responses (Lines 225-227), while the short-term component captured the diurnal responses. I disagree that LOESS can separate model internal variability from aerosol impacts. LOESS is just a statistical tool. If it could filter out model internal variability, scientists wouldn't need to run hundreds of years of simulations or conduct multiple ensemble simulations to minimize model uncertainties. Aerosol signals can accumulate and interact with the atmosphere. While I understand the limitations of 1-month simulations in this study due to computational resources, the authors didn't exploit the existing model results for deep analyses. Most of the analyses are superficial and lack depth and evidence. Instead of relying on these statistical analyses, I suggest the authors delve into model physics and provide a process-level analysis of how global convection-

permitting simulations change or validate our understanding of aerosol-cloud and aerosol-radiation interactions.

We thank the reviewer for this comment but disagree with all of the points raised. We believe our method and analysis are wholly appropriate, robust, and provide original evidential insight and conclusions that are supported by previous studies.

We do not aim to reproduce and separate the full natural variability associated with our perturbation as this is impossible with our fixed SST configuration and relatively short timescales. However, this is not the aim of our study nor the timescale we are focusing on. Instead, our study implicitly focuses on the 'rapid adjustments' of convectively active regions to aerosol perturbations, which is a very different question from studies that aim to capture the full climate response to anthropogenic aerosols. For example, Shipeng et al. (2021) ran 100 year simulations with a mixed-layer slab ocean configuration in order to quantify both 'fast' and 'slow' responses to idealized aerosol perturbations.

From previous studies we know that these rapid adjustments occur within the diurnal cycle, so we expect our response to contain a 'repeatable' signal on a diurnal time scale. Most time series decomposition methods are rigid and not designed for a signal that varies day to day - so inappropriate for a background meteorological state that will vary day to day. The LOESS method allows for these variations and provides an excellent tool for this study.

> From Dokumentov and Hyndman (2022): *"Existing time series decomposition methods are designed for monthly and quarterly data with few tools available for more frequent data. The seasonal-trend decomposition using Loess (STL) procedure of Cleveland et al. (1990) is the only widely available decomposition tool for data observed more frequently than monthly."*

The short-term component from LOESS provides the diurnally varying response throughout the time series. This fluctuates around a value of zero so misses any component of the response that is persistent. Our method attempts to extract this from the LOESS long-term component that will implicitly contain any model response that is not purely due to the aerosol perturbation as we discuss in the manuscript (e.g internal variability). We do not claim this is a perfect extraction and state this in the manuscript (we make this clearer in the revised manuscript - see below).

Though the technique is often referred to differently, we are not the first to use LOESS to decompose responses in the climate system. The power of this statistical decomposition tool (and other similar methods) has been well demonstrated by other studies focusing on the climate, e.g., Deng and Fu, 2019; Carslaw, 2005; Verbesselt et al., 2010; He et al., 2022; Liu and Zhang, 2024; Zhou et al., 2015; Cleveland, 1979; da Silveira Bueno et al., 2024; Papacharalampous et al., 2018; Quan et al., 2016; Jaber et al., 2020; Rabbi and Kovács, 2024; Moradi, 2022; Deng et al., 2015.

We have revised the manuscript to address the reviewer's concerns. First, throughout the manuscript (abstract, introduction, conclusions) we now clearly state that we are focusing on the rapid response due to the aerosol perturbation. Second, we have revised Sect 2.4 (Temporal decomposition of regional response) to clarify that the isolation of internal variability is approximate. We also include more description of the LOESS method, its applicability to our research question, and include citations to several previous studies. Finally, we acknowledge the limitations associated with the temporal decomposition method in the conclusions.

In Sect. 2.4:

We attempt to isolate the responses due to the aerosol perturbation from internal variability and noise by temporally decomposing…

"LOESS is a statistical decomposition tool that can be applied to extract responses occurring on relatively high frequencies (e.g. diurnal) and has been used in previous climate-focused studies (e.g. [citations…])."

"...may represent an important aerosol effect, hence we attempt to recapture this using a second application of the decomposition tool"

"This method assumes that any internal variability is evenly distributed around the time-independent response, which may not be true, but provides a reasonable approximation and should capture regions where strong persistent responses occur."

In the conclusions:

"In an effort to isolate the aerosol impacts from internal variability, we subset the globe"

"Future studies should also consider building on the temporal decomposition method (Sect. 2.4) as not all internal variability can be isolated from the aerosol-driven response. The method assumes that mean internal variability during the time series is equal to zero; whilst this may be true on very long time scales (years to decades) it is unlikely to be the case over our simulation duration. The method additionally assumes that the persistent response due to the aerosol perturbation is independent of time. In reality, this component may increase or decrease during the simulation due to local or non-local feedbacks between clouds, the surface, and the thermodynamic properties of the region. This could be explored in future studies with longer simulations."

Secondly, the authors divided the globe into 288 15°×15° regions and treated the global simulation as a sum of 288 regional simulations. While this approach is acceptable, it has limitations. The authors should discuss the limitations, especially the interactions between nearby 15°×15° regions. The transport effect should be significant in the 30-day simulation.

We did not perform 288 separate simulations, rather we took the outputs from the global simulations and subset them into 288 defined regions. Using our method, the mean global response will be the same whether it has been calculated from variables on the native grid (~5 km) or from the 15°×15° grid. The regional subsetting is performed in order to encompass a large enough number of native data points to maximise the use of the LOESS temporal decomposition method.

Unlike traditional regional-scale simulations, our configuration implicitly allows neighbouring regions to interact with each other. Thus, the transport of energy, moisture, etc to neighbouring regions (and further afield) can be seen to be included as boundary conditions. The cloud response due to the aerosol perturbation in one region will indirectly influence the response in the neighbouring region, and so on. We view this as a benefit as it permits non-local regional responses to an aerosol perturbation, aiding in the realism of the response.

In the revised manuscript we have clarified how the '288 equivalent simulations' were achieved by taking the global outputs and then subsetting into regions:

"To study aerosol impacts on the global scale we subset the outputs from the global simulations into 15°×15° regions, producing the equivalent of 288 regional-scale simulations running for a 30 d period. With this method, the regions can interact with each other, and any regional aerosol response is transported to neighboring regions."

"In a global-scale analysis we subset the global simulation outputs into 15°×15° regions, producing the equivalent of 288 regional high-resolution simulations that can interact with each other."

Thirdly, as mentioned in the first major comment, the analyses lack depth and robust evidence. This issue was raised in the first round of review but was not significantly improved in the revised manuscript. The four 1-month simulations are complete, and I suggest the authors think carefully about how to exploit the existing simulations for a comprehensive analysis.

Our analysis addresses our objectives and provides an original perspective into the variable role of aerosol in the climate system. The global-scale analysis is consistent with literature and the process-level analysis supports our conclusions, which are wholly in-line with literature from the community. We acknowledge in the manuscript that there are some limitations to the study design, but these are unlikely to greatly affect our core conclusions. This study has provided an excellent starting point for us to include interactive aerosols in the ICON model and other kilometre-scale modelling configurations and will help shape the design of future studies and model intercomparison projects.

Fourthly, some sentences are difficult to understand. I had to read the manuscript several times to grasp (or guess) the authors' intentions. If the authors plan to revise the manuscript, language improvement is necessary.

Following this, and a similar suggestion from Reviewer 1, we have revised the manuscript to improve the language and readability.

Since most of the analyses are based on the LOESS decomposition, I won't comment further on those results, even though I agree with some conclusions. Please find below additional detailed comments.

Title: How about "Regional variability of aerosol impacts on …"? "Regional-scale aerosol impacts" is misleading.

We agree with the reviewer and have revised the title as suggested.

Lines 31-43: Please consider reorganizing these sentences.

As suggested by the reviewer, the third, fourth and fifth paragraphs in the introduction (which include lines 31-43) have been reordered and revised to provide a more logical story.

Lines 65-67: Your simulations seem not to be able to reproduce the observations, as shown in Figure 4. Is my understanding correct?

The sentence in question is "[Sato et al. 2018] found that using an explicit representation of cloud microphysics on a global scale produced a negative LWP-AOD relationship, in agreement with satellite observations, that was not replicated in a coarser global model."

We use this study in the introduction as an example of how model resolution can impact the representation of cloud microphysical processes, rather than to characterise the relationship between LWP and Nd. The annual-mean relationship between LWP and Nd in liquid topped clouds (as shown in Sato et al. (2018)) is known to be strongly nonlinear (Gryspeerdt et al., 2019) and difficult to quantify from remote satellite observations (Jia et al., 2022; Grosvenor et al., 2018; Quaas et al., 2020). Gryspeerdt et al. (2019) and others have found that at low values of Nd the dLWP/dNd relationship is likely positive, and at higher values of Nd the relationship is likely negative. A linear trend between extreme ends of the Nd distribution hides this complexity and does not consistently result in a negative trend as observed by Sato et al. (2018). The magnitude and sign of the trend is sensitive to the satellite product (Gryspeerdt et al., 2019; Grosvenor et al., 2018), the region of interest (Michibata et al., 2016), and meteorological variables (Michibata et al., 2016; Sato et al., 2018). Additionally, as we conclude in our manuscript, remote sensing observations may misrepresent the daily-mean response of clouds to aerosol perturbations as they only observe a short time window in the diurnal cycle, as demonstrated by Figure 10.

Therefore, the observations of the LWP-Nd relationship presented by Sato et al. (2018), and their comparison method, are not directly applicable to our simulation but do provide an interesting point of discussion. The most relevant clouds to compare are those in the marine stratocumulus region of the SE Atlantic. Although we observe a diurnal cycle in the LWP response to the aerosol here (Figure 4c, Figure 10f) it is always positive when the persistent effect is included (Figure 6b and 6c). This is consistent with Michibata et al. (2016) but inconsistent with Sato et al. (2018).

Gryspeerdt et al. (2019) suggest that the contrasting LWP-Nd relationship at low vs high Nd is related to the switch from precipitating to non-precipitating clouds. Sato et al. (2018) also show they were able to obtain a negative LWP-Nd relationship when the representation of precipitation was improved. This suggests our LWP response may be sensitive to the representation of precipitation and its link to Nd, which is simplified in our model (autoconversion).

We include the following in Section 3.2.1:

“The positive relationship between ΔLWP and $\Delta N_{d,cld}$ in the SE Atlantic is consistent with remote sensing observations from Michibata et al. (2016), but inconsistent with those from Sato et al. (2018), and may be sensitive to the representation of the warm-rain process (Gryspeerdt et al., 2019; Sato et al., 2018, Terai et al., 2020); we revisit this in the conclusions.”

And include the following in the Conclusions section:

”Additional sources of uncertainty arise from the cloud microphysics scheme and unresolved convection. The choice of cloud microphysics scheme and representation of cold-phase processes have been shown to impact the sensitivity of convective clouds to aerosol (Heikenfeld et al., 2019; White et al., 2017; Sullivan and Voigt, 2021; Marinescu et al., 2021), while the representation of the warm-rain process and its link to aerosols have been shown to be important for ACI impacts on warm-phase clouds (Gryspeerdt et al., 2019; Sato et al., 2018, Terai et al., 2020).”

Line 85: What is JSBACH?

This acronym has now been defined as follows:

“.. the Jena Scheme for Biosphere Atmosphere Coupling in Hamburg (JSBACH) dynamic vegetation model”

Lines 107-109: I am still confused about your definition of biomass burning. Do you consider it an anthropogenic source? If not, the selection of September due to its remarkable biomass burning does not make sense since your study focused on the impact of anthropogenic aerosols. It doesn’t hurt your experimental design, but please describe the model setup and reasoning as accurately as possible.

Lines 128-131: Again, it is confusing whether biomass burning is considered natural or anthropogenic sources in your study.

Lines 135: What do you mean by the natural and anthropogenic contributions of the biomass burning plumes?

Figure 1 and Lines 187-188: It seems that biomass burning is considered anthropogenic sources in this study. I must disagree with that. It would be better to clarify this point and use more accurate wordings throughout the manuscript to distinguish the PD and PI experiments.

In our simulation we assume that anthropogenic activity contributes to emissions of absorbing aerosol, which are represented in MACv2-SP. This is consistent with global databases (van der Werf et al., 2017), CMIP5 and CMIP6 inventories (Lamarque et al., 2010; van Marle et al., 2017), and observations (e.g., Abatzoglou and Williams, 2016; Knorr et al., 2016). The magnitude is uncertain (e.g., Hamilton et al., 2018; Lauk and Erb, 2009) and we use a value at the higher end of this uncertainty in order to maximise the signal.

In this study, we are focused on understanding the regional variability of the role that idealized aerosol perturbations have on clouds and climate, which we frame within the context of anthropogenic activity. We have revised the manuscript to make this point clearer. The aim is to make the reader aware that we are focused on the response of clouds and climate to an idealized aerosol perturbation, for which we use a present-day vs pre-industrial comparison. We have included the following sentence at the end of the introduction:

"We analyze the response of clouds and the thermodynamic environment to an aerosol perturbation by contrasting simulations using aerosol representative of the pre-industrial era with aerosol representative of the present-day."

In the rest of the manuscript, we refer to the change in aerosol as an aerosol perturbation (as opposed to anthropogenic aerosol).

Lines 117-118: What do you mean by non-interactive and spatially invariable but temporally variable? In Line 95, you mentioned that Nd follows a predefined vertical profile. If aerosol concentrations change, will Nd change? How about the vertical profile shape of Nd?

This section has been revised for clarity.

Lines 125-128: Do you mean the 3D fields of aerosol extinction are calculated based on the nine plumes of aerosol concentrations and optical properties?

The lines mentioned are: *"MACv2-SP, described in full by Stevens et al. (2017), provides the model with 3D fields of aerosol extinction that are predefined at the beginning of the simulation. The fields are represented as nine plumes spatially consistent with the dominant sources of global anthropogenic aerosol emissions. Each plume is characterized by parameters that control its horizontal and vertical distribution, aerosol concentration and optical properties, annual cycle, and year-to-year variations."*

For clarity, we have rewritten this as suggested by the reviewer:

"Anthropogenic aerosol perturbations are represented using MACv2-SP, described in full by Stevens et al. (2017), which provides the model with 3D fields of aerosol extinction that are calculated for nine predefined plumes of aerosol concentrations and optical properties. The plumes are spatially consistent with the dominant sources of global anthropogenic aerosol emissions, and each is characterized by parameters that control its horizontal and vertical distribution, aerosol concentration and optical properties, annual cycle, and year-to-year variations."

Line 140: Please explain how the change in the ratio of natural to anthropogenic aerosols can provide a stronger signal if you only change the anthropogenic aerosol concentrations from PI to PD? The current model description is confusing, and I don't understand the logic.

We have revised the manuscript to provide clearer information on how the two sources of aerosol (natural and anthropogenic) are represented, and how we enhance the anthropogenic contribution from the biomass burning plumes in MACv2-SP.

Line 163-164: No. Figure 1 shows the overestimation of Nd compared to the annual climatology.

We have revised our observational climatology to ensure we are making a like-for-like comparison and have expanded the N_d climatology in Figure 1b. In the Grosvenor et al. (2018) manuscript, N_d was shown over oceans only, but the authors also provide data over land in the supporting information. This provides a better test for our aerosol representation as the MACv2-SP plumes are centered over land (where primary aerosol emissions occur), not the ocean. We used this alternative dataset and removed grid boxes with relatively large uncertainty - where the number of days with a successful retrieval over the 13 year time series was less than 50. The revised climatology is shown below in Fig. R1 (duplicated from comment from Reviewer 1).

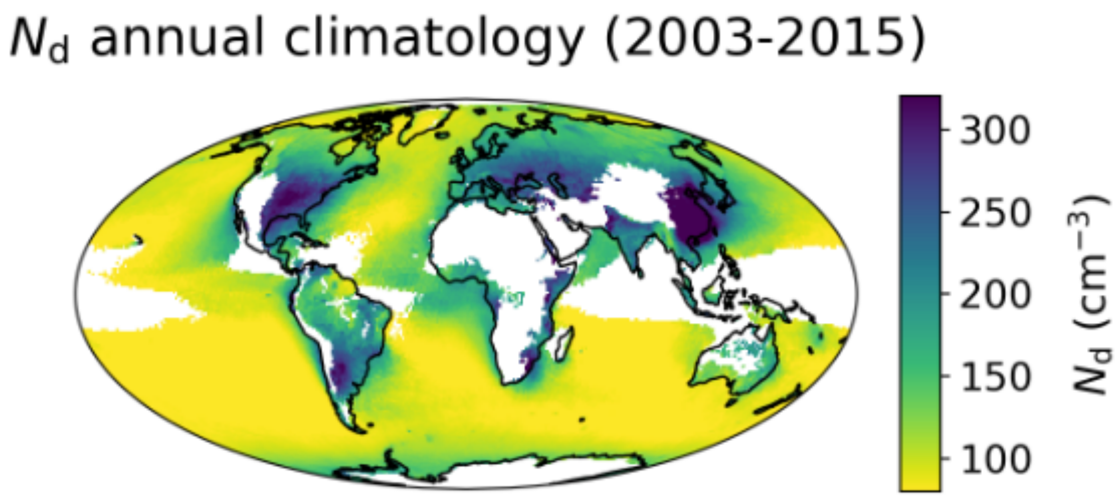

Figure R1. Revised annual mean N_d climatology from Grosvenor et al. (2018). Blank regions show grid boxes where there were fewer than 50 retrievals in the 13 year time series.

A statistical comparison to the climatology using the new and default parameters is shown in Table R1 below (also duplicated from previous comment). Using the more expansive N_d climatology has increased the correlation coefficient from 0.34 (oceans only - unconstrained) to 0.57 (all available grid points - constrained to > 50 data points). This value still demonstrates some discrepancy between simulation and observations but provides more confidence to the reader. We also stress, as we do in the manuscript, that this is representative of the spatial distribution and magnitude of anthropogenic aerosol, and appropriate for our study.

	Herbert et al. (2021) parameters	Default (Stevens et al., 2017) parameters
Root mean square error	49.5	70.6
Mean absolute error	34.0	47.3
Normalized mean bias	-0.12	-0.35
Correlation coefficient	0.57	0.42

Table R1. Statistical tests between observed N_d from Grosvenor et al. (2018) and simulated N_d using the MACv2-SP plume model.

The revised climatology has been included in the revised manuscript, and the following text has been included in Sect. 2.3:

"A comparison between simulated and observed N_d yields a root mean square error (RMSE) of 49 cm^{-3} and a correlation coefficient of 0.57 (the default parameters a_N and b_N yield an RMSE of 70 cm^{-3} and correlation coefficient of 0.42). The discrepancy is in part due to high simulated values over biomass burning regions that are not reflected in annual mean observations, but also due to regional variability that MACv2-SP does not capture (e.g., North America)."

Figure 1b has also been updated to Fig R1 above and the caption has been expanded to describe the new method:

"... and (b) annual mean N_d for cloud tops < 3.2 km (2003 – 2015; only showing grid points with 50 successful retrievals) estimated by Grosvenor et al. (2018)..."

We give reasons for their discrepancy and provide reasoning as to why this isn't a primary concern in the study.

Line 168: I guess Nd,cld is three-dimensional. So you enhance Nd,cld everywhere throughout the vertical column with the same factor. Is my understanding correct?

Yes this is correct. We have rewritten the paragraph to make this clear. We have also emphasized that this produces an idealized distribution of Nd.

"In the default ICON setup, the microphysics scheme uses a predefined value for the cloud droplet number concentration ($N_{d,cld}$) that is spatially invariable and constant in altitude. We use this for our PI distribution of $N_{d,cld}$, which we set to 80 cm^{-3}. We represent ACI effects in the microphysics scheme using the ACI scaling factor f_N from MACv2-SP, as calculated above. Applying f_N to the pre-industrial distribution of $N_{d,cld}$ provides an idealized present-day distribution that is spatially consistent with the anthropogenic contributions in the MACv2-SP plumes."

We also highlight this limitation in the conclusions:

"*The idealized representation of aerosol and N_d in this model has helped identify important process-level interactions and provides a platform for future studies using realistic aerosol perturbations. The use of non-interactive aerosol may mask important feedbacks and processes including the impact of clouds and precipitation on the spatio-temporal distribution of aerosols, changes to the surface properties and energy fluxes, and turbulence that would influence emissions and aerosol removal processes. Changes in aerosol concentrations would also affect N_d concentrations and vertical profiles.*"

Lines 179-182: Figure 1 can't distinguish anthropogenic from natural aerosols. It would be better to add a supplemental figure showing the anthropogenic and natural aerosol emissions to support this statement.

This has now been included in the supporting information (Sect. S1 and Fig. S1) and referenced in Sect. 2.3.

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