We thank the anonymous reviewers and the editor for the time and attention they dedicated to our manuscript "Introducing aerosol-cloud interactions in the ECMWF model reveals new constraints on aerosol representation". We also appreciate the recognition of relevance of the motivation of our work and are happy to submit a significantly revised version of the paper stemming from the reviewers' concerns. We updated the entire optimisation system under various aspects, above all increased resolution in space (from 3x3 to 1x1 degrees) and time (from 6- to 3-hourly data for each sampling date). These allowed us to refine the model data selection criteria to better match those of the satellite retrievals, so that in the new system only daytime datapoints are considered, and that at the increased grid resolution the ice phase can be excluded more effectively. In addition to this, a diagnostic treatment of vertical velocities is introduced, and the activation lookup table now includes also two additional dimensions for mean and standard deviation vertical updraft. Below follows a detailed response to each comment. We also largely revised the structure of the paper, to improve readability and clarity. The result section has been split into separate sections for the results, the wet scavenging sensitivity experiments, and the discussion. We also simplified the setup of the sensitivity experiments by reducing the number of changes and making the attribution of effects more straightforward.

Reviewer 1 - Comment 1

1. Model description clarity, effective radius, autoconversion

"The model/simulation description and configuration are not very clear. The study aims to "introduce aerosol-cloud interactions," but it does not mention: a) whether and/or how the computed Nd values will affect cloud droplet effective radius and autoconversion calculations; and b) whether the modified cloud properties/lifetime will provide feedback to radiation or meteorology. For the results discussed, it is often difficult to determine whether they are from offline calculations or online simulations."

Reply: We acknowledge that the description of the model, especially concerning the IFS cloud scheme, was insufficient. We updated the manuscript to better describe that N_d interacts with the IFS radiation scheme but not yet with the cloud physics, with reference to the IFS documentation for extra details on the cloud scheme.

We make this point explicit within the abstract:

> aerosol validation and assimilation, only weakly constrains aerosol number concentrations relevant for cloud formation. We introduce a physically informed representation of aerosol-cloud interactions (ACI) in the European Centre for Medium-Range Weather Forecasts (ECMWF) into the ECMWF Integrated Forecasting System (IFS)to simulate the activation of aerosols into cloud droplets driven by aerosol concentrations from , restricted to aerosol activation and the first indirect effect, and use it as a diagnostic tool to relate aerosol properties to cloud observations in the Copernicus Atmosphere Monitoring System

And detailed the rationale at the beginning of the Method section:

> **4 Method**
>
> We constrained aerosol numbers This study on ACI considers only the first indirect aerosol radiative effect, excluding any direct impact on cloud lifetime. This choice focuses on the activation pathway, without the introduction of additional feedback mechanisms, and reflects the more limited understanding of the effect of aerosols on precipitation compared to the first indirect and the semi-direct radiative effect (Stier et al., 2024). Moreover, if lifetime effects were active, the optimisation problem would no longer be well-defined, as N_d changes would directly affect precipitation and cloud evolution, while the optimisation relies on stored cloud fields. This section presents how we constrain aerosol number concentrations for the bulk IFS representation

And repeated in the conclusions:

> **8 Summary, outlook and conclusions**
>
> In this studywe showed that , we introduced an explicit representation of aerosol-cloud interactions (ACI)in a weather model can provide novel constraints on modelled aerosol processes. To do this, we introduced a lightweight aerosol activation scheme into the , limited to aerosol activation and the first indirect effect, in the ECMWF Integrated Forecasting System (IFS)used to implement , enabling cloud droplet number concentrations (N_d) to be used as an additional aerosol diagnostic. By combining

2. Lacking evaluation of CAMS aerosol mass fields

“The optimization only considers the impact of aerosol size distribution (or r_{med} changes on N_d, assuming that the aerosol mass fields and thermodynamical fields for activation calculations and the sampling/averaging are "perfect." The authors did not provide any evaluation of these fields or relevant references. For example, do CAMS aerosol forecasts have known substantial biases and uncertainties over regions with stratiform warm clouds? What is the uncertainty associated with sampling errors? Without considering these aspects, the optimization could compensate for systematic biases in these fields through unrealistic size distribution adjustments.”

Reply: The aerosol model is routinely evaluated by the CAMS community and its description, together with evaluation against a wide range of observations can be found in Rémy (2022), where a good performance of the global fields is assessed (and which we now cite); aerosols are evaluated also in CAMS reports several times a year and their performance (with and without assimilation) can also be assessed with tools like AEROVAL (https://aeroval.met.no/pages/evaluation/?project=cams2-82). We included these references in the paper:

> period 2003-2020, and 2003–2020. Evaluations of the CAMS aerosol system (with and without assimilation of observations) can be found in Rémy et al. (2022) and in the CAMS validation reports released periodically (see e.g. Blake et al. (2025)); near real-time validations are also publicly accessible using tools such as Aeroval (https://aeroval.met.no, accessed April 2026). Simulated aerosol fields were stored on 16 pressure levels between 10 hPa and 1000 hPa. The ; the 11 pressure levels below

Moreover we included global evaluation of satellite AOD and AE for the default and optimized size distribution definitions in Figure 5. As discussed in the paper, we are aware that such fields are certainly not perfect and incorporate many unconstrained aspects, concerning speciation, total mass, physical and optical properties, sizes and shapes, and 3D spatial distribution. We are also totally aware that the optimization does tend to compensate for associated systematic errors. This is now stated more clearly in the Method section:

As a result, a collection of optimised median radii for each species x and year y is obtained, $r_{\mathrm{med},x,y}$. We finally define the optimal PSD as the one using the optimal median radius $\overline{r_{\mathrm{med},x}} \equiv \mathrm{med}_y(r_{\mathrm{med},x,y})$, i.e. the median across the 18 years of the optimized optimised median radii. Within this framework, the resulting optimised aerosol PSDs are constrained by the cloud-top N_d observations and represent an effective description of aerosol number consistent with these observations; their realism is conditioned by non-represented processes (e.g. diagnostic representation of N_d and absence of prognostic droplet microphysics) and aerosol biases (e.g. speciation and vertical structure), and is therefore evaluated *a posteriori* through independent observations of aerosol optical properties.

And at the beginning of the Discussion, with a new subsection addressing the effectiveness of the optimisation

deals with the systematically low N_d at mid- and higher latitudes in the Southern hemisphere, where the optimisation is unable to bring improvement, and how the outcome of the sensitivity tests compare with the optimisation results. In both cases, results from the different setups (InCloud, InCloudNoBC, ClBase3), combined with independent observations, are used to clarify to what extent each optimal PSD can be interpreted as physically plausible, and when it compensates for non-tuned aspects such as speciation, ageing or poor mass concentration representation.

7.1 Limitations and effectiveness of the N_d optimisation

While the magnitude of reduction of $\mathcal{L}$ by only 13% may raise doubts about the effectiveness of the optimisation, it actually indicates that the prior (CAMS default) PSDs yield N_d values already close to the optimisation target. The intrinsic limits of the optimisation lie in the prescribed spatial aerosol patterns from the model fields, and the restricted number of free parameters (the PSD r_{med} for five species). This is illustrated by the ClBase3 optimisation setup, where the prior N_d prediction has much larger errors throughout the tropics; in this setup, the improvement brought by the optimisation is significantly stronger, with a reduction of $\mathcal{L}$ by 21%.

We aimed to make it more explicit throughout the manuscript that it is indeed within the scope of this work to show how those biases whose patterns cannot be described by the few degrees of freedom of the optimization procedure (one global value of r_{med} per each species) are automatically highlighted in the resulting optimized setup. In particular, the biases addressed in this study, such as the Southern Ocean, might be undetected in the standard setups used for validation, due to the lack of reliable observations over open seas and, at the same time, they are insufficiently compensated by the tuning procedure. In the case in point, we concluded that these biases reflected issues related to the aerosol population found inside (and directly below) the cloud over the region, which gave insights on which processes are the most promising to improve the aerosol model in this sense. However, this work is not the last word on the matter - applying the optimization has highlighted areas in both size distribution assumptions and in the CAMS mass concentrations, and the intention is that future work builds on this paper to improve both mass distributions and size distributions simultaneously.

3. Critical model/retrieval sampling mismatch

Critical model/retrieval sampling mismatch. The comparison between model-derived and satellite-observed Nd may suffer from sampling inconsistencies that invalidate the optimization results

The reviewer structured their concerns into the following points (here reported in bold):

a) *different cloud detection methods: ERA5 model diagnostics vs. satellite radiance retrievals define "cloud top" differently.*
Reply: Cloud top properties from MODIS are inferred from brightness temperature measured in some infrared channels. The retrieved temperature is representative of the top layers of the cloud and is typically within 1 cloud optical thickness (τ) in the IR (e.g. 0.72 reported in Wang, 2014). From Mitchell (2000) we get that the extinction efficiency for infrared wavelengths in liquid clouds is between 1 and 2. Since the offline τ estimate is done assuming extinction efficiency 2 (valid in the geometric optics limit), we chose the threshold in visible τ = 2.
We expanded the Method section to include this and the quantitative impact of relevant sensitivity tests to the choice of τ threshold

> water is below 0.5. By doing this, we aim at selecting data points where both the model and MODIS see Optical thickness is diagnosed offline assuming a constant extinction efficiency of $Q_{\text{ext}} = 2$. We chose the cloud-top threshold for consistency with MODIS cloud-top properties being typically within $\tau \approx 1$ in the infrared (Wang et al., 2014a), given that the extinction efficiency for wavelengths of ≈ 10 µm in liquid clouds is between 1 and 2 (Mitchell, 2000). Stratocumulus clouds also have optically thick and geometrically sharp cloud tops, spanning several units of optical thickness within one model level, which makes the results relatively insensitive to the precise choice of the optical thickness threshold for cloud-top identification. For completeness, we tested the sensitivity to cloud-top threshold values of $\tau = 1$ and $\tau = 7$, consistent with bounds provided by correction formulas in Grosvenor et al. (2018a) for r_e retrievals. This affected cloud-top height location by less than 30 hPa for the thickest clouds, and produced variations within 0.7% in the global loss function (see below for its definition). Within a

b) *spatial resolution mismatch: 3°×3° model data (aerosol concentrations and meteorological fields) are used to calculate Nd and compared with MODIS data - although MODIS data are also regridded to a 3x3 grid, they are aggregated and averaged from finer resolution, so they represent vastly different sampling volumes.* (Includes also question c)
Reply: We welcome this concern and agree therefore we have updated the optimisation system to work at a 1x1 degree resolution, while the sub-sampling frequency for each date was increased from 6- to 3-hourly data, and only daytime points are considered, to eventually exclude dependencies on the daily cycle (cloud height, vertical updrafts...):

> For each data stream we select within each month every fifth day starting the third day of the month; for each day, we take the use 3-hourly data (00, 03, 06, 12 and 18 UTCtime-steps..., 21 UTC). All fields are interpolated on a rectangular 3° × 3° grid. In order to find CAMS aerosol concentrations at the cloud level, we consider only to the same 1° × 1° grid of the N_d retrievals datasets.
>
> For each column, only points with local time between 09:30UTC and 15:30UTC, to reflect the typical daytime overpasses of the Aqua and Terra satellites. Only cloudy model grid-boxes are then retained, defined as those with at least 10% cloud

For the future, comparison could potentially be carried out an even higher resolution against level 2 instantaneous overpass data. This could be surely beneficial to verify the ability of the model to simulate sharp changes in aerosol fields, e.g. in the case of strong aerosol emission events like wildfires. The optimization presented in this work is, however, performed against monthly-mean satellite observations, which allow us to catch seasonal aerosol and cloud signals, but is not meant to assess the ability of the model to exactly match the instantaneous N_d values. From this standpoint, the optimization relies on a statistical comparison of simulated and observed N_d values, with an implicit low-pass filtering at timescales shorter than 1 month. This is now also made clearer in the text:

> While aerosol and cloud data are spatially co-located, only monthly mean values are compared, so that $\mathcal{L}$ can be seen as a statistical evaluation of the simulated N_d. This implies that the optimisation targets large-scale and seasonal N_d signals, with variability on monthly or longer timescales. As a result, a collection of optimized the sensitivity to the co-location in time and space of cloud fields and satellite overpasses is reduced. At the same time, by considering a smaller number of samples within each month we can substantially reduce the amount of data processed, keeping the procedure manageable.

c) *temporal sampling bias: model data (4 times daily, every 5th day) vs. actual satellite overpass times.*
Reply: (addressed in answer to question b)

d) *vertical sampling inconsistency: model-diagnosed cloud levels vs. satellite-retrieved cloud properties may sample completely different atmospheric layers. The current study lacks uncertainty estimates related to these issues.*
Reply: Representativity of the aerosol layer constitutes an important challenge to our method but must also be related with the low vertical resolution of used data on pressure levels as pointed out in section 4.3 (Model data selection), which is on average 1.5km throughout the troposphere. However, since we are concerned with aerosol impacts on liquid clouds, we are typically dealing with boundary-layer clouds, and the vertical difference in sampling between model and satellite is small; in these cases, the aerosol is likely to be well mixed within the boundary layer. Moreover, deep cumuli are systematically excluded by the procedure mostly thanks to the cloud-top physical requirements for temperature and phase. We added more details in the Method section:

> Simulated aerosol fields were stored on 16 pressure levels between 10 hPa and 1000 hPa. The ; the 11 pressure levels below 100 hPa levels have thicknesses ranging between ≈ 640 m and ≈ 2300 m, with the highest resolution found in the lower troposphere and lowest resolution in the mid-troposphere. Such low vertical resolution is clearly suboptimal for the purpose of extracting accurate aerosol concentrations at cloud level; however, recycling these data allows using these data enables us to get useful insights on the current state of the model climate and the potential of the tuning procedure, without needing to re-run a rather computationally-expensive simulation. optimisation procedure, which we also expect to partially compensate for resolution-driven inaccuracies.

and:

> **4.5 Optimization The offline optimisation procedure**
>
> Aerosol mass concentration fields are interpolated at the representative cloud top At each stored time and for every column of the domain, we interpolate aerosols to the identified cloud-top level p and converted to calculate aerosol number concentrations using equation 2. The Equation 2. Given the relatively low vertical resolution of the aerosol dataset, the extracted population at cloud level represents a vertically smoothed average of actual simulated populations above and below the cloud level. The

4. Activation of aerosol population at cloud top - physical inconsistencies

"Physical inconsistency in process representation considered for optimization (Section 4.3). If I understand it correctly, the authors extract aerosol concentrations at "cloud top" and apply a 1 m/s updraft velocity to simulate activation at this level using the lookup table. I assume this approach considers that Nd retrievals are for "cloud top" only, but the method contradicts basic cloud microphysical principles. In reality, CCN activation occurs at cloud base during initial adiabatic ascent where supersaturation develops, and the resulting droplet population is then transported vertically through the

cloud. Nd satellite retrievals often assume adiabatic conditions, where Nd is considered constant throughout the cloud. Additionally, cloud tops typically experience subsiding air masses, entrainment of dry air, and near-zero or negative vertical velocities, so the 1 m/s (upward) vertical velocity assumption at cloud top is very likely unrealistic for most of the time. Aerosol populations at cloud top have been modified by scavenging, entrainment, and chemistry, making them fundamentally different from the original CCN population (consistent with results shown in figure 10). The authors seem to have realized this issue, as indicated by the comparison of InCloud and ClBase3 results in Table 3 and Figures 4 and 10."

Reply: We welcome the call of the reviewer for clarity and consistency on the aerosol activation microphysics. This limitation of the presented setup reflects the choice to aim for a flexible setup that allows the use of relatively long time series of global observations. As reported in the paper, we experimented picking aerosol mixing ratios at different levels within the cloud, the extreme being picking aerosols below the cloud - while this has an impact on the amount of diagnosed sea salt CCN, overall results were quite similar, which may be partially attributable to the intrinsic smoothness of the aerosol fields across the vertical levels, as discussed above. Concerning the choice to pick aerosols at the cloud level to get diagnostic N_d estimates, this is the same approach adopted in the Jones, 2001 scheme of the Met Office climate model, where N_d is, like in our setup, a purely diagnostic quantity as described in West, 2014. The experiment in which we picked aerosols below cloud base was intended to address the specific issue of missing aerosols inside high-latitude clouds, yet besides the modest (but insufficient) impact at high latitudes, we got limited improvement elsewhere and no change in qualitative terms, with a persistence also of the strong bias over Africa as shown in the paper. We added an explanation of this choice in the text

> Section 5.2), and a third one using aerosols found three levels below the cloud base (ClBase3, Section 5.3). Although CCN activation mainly occurs at cloud base, the model does not represent aerosol as explicitly dissolved within cloud droplets. The aerosol at cloud top has nevertheless been transported there by the same turbulent, mixing and advection processes that transport cloud water. In that sense, the aerosol population co-located with cloud droplets at cloud top (InCloud) provides a first-order diagnostic representation of the aerosol consistent with cloud-top $N_d$retrievals are known to suffer from an optical penetration bias due to the assumption that the spectrally-retrieved r_e values are representative of the PSD of cloud droplet at cloud top, which leads to overestimated retrieved , despite activation occurring at cloud base. The InCloudNoBC and ClBase3 setups are presented to check the robustness of the procedure to the CAMS BC representation and to the aerosol vertical pro-

Regarding the choice of a fixed updraft velocity of 1 m/s, we agree with the reviewer's concerns and have substantially improved the estimate of updraft velocity, as described in our response to Reviewer 2 (comment 3) – see below.

The title of the paper

"If the goal is really to show the introduction of ACI in IFS helps to constrain the aerosol representation, a more comprehensive evaluation of the aerosol properties is needed (e.g., evaluation of aerosol size distribution using in-situ data). In my opinion (and as the authors discussed in the introduction), the value of this work is more on providing a simplified but practical treatment of Nd prediction and ACI representation in the IFS model, which will allow IFS (with CAMS) to consider the impact of aerosols on clouds in the future."

Reply: We agree that it is worth extending the work by bringing in more observations, especially from in-situ measurement sites or specific campaigns. This paper is however quite long already, and we consider investigation in this sense (combining aerosol indirect effects and in-situ aerosol measurements) certainly worth being done and eventually published in the future. This is indeed pointed out in the abstract and in the conclusions, where we provide outlooks and suggestions in this direction stemming from the results of this work. We also believe that the title accurately describes the scope of the paper, which is to introduce, as the reviewer correctly puts it, a simplified but practical treatment of Nd to illustrate that a new set of observations (in this paper N_d and all-sky SW fluxes), can be now brought in to constrain the CAMS aerosol system. Moreover, we present two cases as a proof of concept to illustrate how our approach can reveal regional deficiencies in the aerosol distribution that should be investigated in future. We made this point more explicit in the introduction:

> collocate aerosols and clouds to accurately assess characterise the atmospheric composition at cloud level. Thirdly, instead of optimizing optimising mass-to-number conversion coefficients, we optimize optimise the underlying particle size distribution (PSDdistributions (PSDs) of the bulk aerosols . By using and evaluate their physical consistency against independent observations of aerosol optical properties. Using this approach, we are also in the position to can also interpret the results of the optimization optimisation in terms of aerosol representation and assess evaluate consistency with other represented processes, such as theirdirect radiative effect. Finally, results of the optimization procedure could be tested with simulations using the IFS the ACI scheme resulting from the optimisation procedure is tested in IFS simulations with prognostic aerosols.
>
> The core question of this study is whether N_d observations can globally constrain aerosol number representations in a system used for operational atmospheric composition analyses and forecasts, and which deficiencies in aerosol processe such constraints reveal. This work has the following structure: Section 2 illustrates describes the mass-bulk representation of aerosols

Punctual comments

We directly adopted most punctual remarks and corrections (well spotted, thanks!) made by the reviewer. Those that required a more articulated response are reported below in bold and followed by our reply.

1) *Page 1, Line 10: "We found that CAMS aerosols allow simulating overall realistic Nd values". Is this conclusion for unoptimized or optimized ASD?*
   Reply: After optimisation. Added clarification in abstract. However, also the prior values were fairly acceptable, and we discuss this explicitly in the new version:

   > **7.1 Limitations and effectiveness of the N_d optimisation**
   >
   > While the magnitude of reduction of $\mathcal{L}$ by only 13% may raise doubts about the effectiveness of the optimisation, it actually indicates that the prior (CAMS default) PSDs yield N_d values already close to the optimisation target. The intrinsic limits of the optimisation lie in the prescribed spatial aerosol patterns from the model fields, and the restricted number of free parameters (the PSD r_{med} for five species). This is illustrated by the ClBase3 optimisation setup, where the prior N_d prediction has much larger errors throughout the tropics; in this setup, the improvement brought by the optimisation is significantly stronger, with a reduction of $\mathcal{L}$ by 21%.

2) *Page 4, Line 93: The direct aerosol effect should also affect the meteorological fields, not only semi-direct effect.*

Reply: Corrected, thanks.

> includes the direct radiative effect of aerosols. Meteorology impacts which is the default for operational NWP. Meteorology affects aerosol optical properties via hygroscopic growth and, in a prognostic configuration, their production, transport and removal. However, feedbacks of aerosol-related processes aerosol feedbacks on meteorological fields are only mediated by influence of aerosol-radiation interactions the impact of the direct radiative impact of aerosols on heating rates(semi-direct effect of aerosols). The IFS .

3) *Page 4, Line 105: Will hydrophobic aerosols be removed by precipitation?*
Reply: No, hydrophobic aerosols are not removed by precipitation. Rephrased in text for improved clarity.

> with water, which are relevant to transport processes and radiative transfer calculations. Hydrophilic species are also removed undergo a removal process when precipitation occurs, with a fixed mass scavenging rate described in ECMWF (2024a) and following Luo et al. (2019). Aerosols in the IFS are represented as

4) *Page 4, Line 115: Does hydrophilic BC have hygroscopic growth and is it considered in activation? Or "hydrophilic" BC only applies to wet removal calculation?*
Reply: Clarified in the text (the "growth refractive" typo is corrected in the non-diff manuscript):

> of the dry particles are unaffected. Hydrophilic OM undergoes hygroscopic growth, while hydrophilic BC is optically identical to the hydrophobic type, which represents pristine soot monomers. In reality, aged BC appears in the form of clusters or chain-like aggregates (e.g. Teng et al. (2019) and references therein) that clump with hydrophilic sulfate (SU)SU, nitrate (NI)

5) *Page 6, Line 144: It seems to me the change in K_{ext} is quite large for certain spices at certain RHs. It would be useful to calculate the difference using the default and optimized r_{med} values.*
Reply: A possible illustration of this effect is depicted in Figure 6, showing that quite a significant impact is attributed to zonal mean additional 0.01 AOD from Sea Salt, peaking over the Southern Ocean 0.02. Contribution from OM is instead negative and peaks in the tropics over South-East Asia, India, Central Africa.

6) *Page 8, Line 165-170: Please provide the references of Q06, G18, and BR17.*
Reply: Done.

7) *Page 10, Line 216-217: How large is the uncertainty associated with these assumptions?*
Reply: We added reference to previous studies, including one evaluating the same parcel activation model:

> being lifted for lifted for a minimum of 100 m at 1 m/s. There is no until peak S_{sat} is reached.
>
> Previous studies found weak dependency of the LUT on atmospheric variables other than aerosol number concentrations. Vertical velocity is known to have a relevant impact on activated particle numbers, especially in regimes with activation process on cloud-base pressure and temperature, especially for warm phase regimes (above 273 K) (Leaitch et al., 1986; Rothenberg and Wang, 2016). We perform simulations for a range of vertical velocities w (updrafts) to precisely describe also regimes

We also discussed more extensively method choices specific to our study in the

Method section:

> Now we can estimate how a finite change in the median radius r_{med} can affect N_d. Let's consider as an example sulfate aerosol (frequently a dominant CCN provider) and a change in its PSD r_{med} from $0.10\ \mu\text{m}$ to achieve this, we firstly implemented a $0.08\ \mu\text{m}$. This will affect N_d by changing both the activation rate α_i and the particle number concentration N_i. The change in α_i derives from r_{med} determining the critical supersaturation (S_{crit}) distribution and thus the maximum supersaturation during ascent. Using results from Rothenberg and Wang (2016), we estimate $|\frac{\Delta\alpha_i}{\alpha_i}| \lessapprox 0.2$. Rothenberg and Wang (2016) report that r_{med} is also the largest controller on activation rates α_i, so that we can neglect from now on the potential effect of changing the geometric standard deviation σ and the hygroscopicity κ. The effect of the r_{med} change can be then estimated using Equation 2 in the approximation of $r_{\text{min}} = 0$ and $r_{\text{max}} = \infty$: $\Delta N_i/N_i \approx \Delta J/J \approx -3\Delta r/r$, so that $|\Delta N_i/N_i| \approx 0.6$. Thus, the aerosol PSD median radius r_{med} exerts the strongest control on N_d through N_i, and secondarily through α_i. This ensures that, to a first order, we can estimate the impact of r_{med} on N_d by neglecting the impact of r_{med} on the S_{crit} curve, i.e. by avoiding new parcel ascent computations and keeping α_i constant. In addition to r_{med}, α_i is controlled by the updraft velocity w (Rothenberg and Wang, 2016), which is the only environment quantity that we will diagnose for estimating N_d.

8) *Page 10, Line 224: The assumed updraft/vertical velocity is pretty large, and is associated with large uncertainties.*
   Reply: This point is similar to point 3 of reviewer 2, and is addressed below.
9) *Page 11, Line 238: T255 should be at \$~80km\$ resolution, instead of \$38km\$? Please double check.*
   Reply: Well spotted thanks!
10) *Page 11, Line 250: Does the MODIS retrieval apply a similar conditional sampling?*
   Reply: Partly, since in the first version we needed to relax the ice water ratio IWR = IWC / (IWC+LWC) criterion due to the very coarse (3x3 degrees) horizontal resolution, where we allowed IWR up to 0.5. With the higher resolution of 1x1 degrees we make the criterion stricter and significantly closer to the satellite filtering

> for the thickest clouds, and produced variations within 0.7% in the global loss function (see below for its definition). Within a cloudy model column, we select liquid-phase stratocumulus clouds . Note that clouds as those where at cloud top $T > 268$ K, and $\text{IWC}/(\text{LWC}+\text{IWC}) < 0.1$ similarly to the observational filtering criteria, but allowing for some ice within the gridbox due to the coarse resolution of the model fields ($3° \times 3°$) , we adopted selection criteria that are more relaxed than those used for the $1° \times 1°$) compared to the level-2 data the N_d retrievals. datasets were derived from (Gryspeerdt et al., 2022b).

11) *Page 12, section 4.3: I assume this approach considers that Nd retrievals are for "cloud top" only, but the method contradicts basic cloud microphysical principles. In reality, CCN activation occurs at cloud base during initial adiabatic ascent where supersaturation develops, and the resulting droplet population is then transported vertically through the cloud. Nd satellite retrievals often assume adiabatic conditions, where Nd is considered constant throughout the cloud.*
   Reply: This was addressed in main point above about physical inconsistencies of the activation of aerosol population at cloud top.
12) *Page 12, section 4.3, formula 7: Why only $N_{d,Q06}$ is considered in the numerator?*
   Reply: We added a remark in the Optimization Procedure section clarifying that our choice is because, being Q06 the least restrictive sampling strategy, it

contains by design the largest number of valid retrievals:

> We use the Q06, G18 and BR17) N_d retrievals averaged to monthly means and re-gridded them to a $3° \times 3°$ rectangular grid on the original $1° \times 1°$ rectangular grid for the period 2003–2020. We use Q06 as the tuning target , while we take the spread across the three datasets to estimate the uncertainty of observations. For each pixel, error bars are defined optimisation target due to its broader spatio-temporal coverage, and define error bars as the full dispersion (maximum minus minimum) of values spread across the three datasets . If data are missing in one of the datasets (because there are no valid retrievals satisfying the

13) *Page 12, Line 285: a brief description of the "Nelder-Mead" algothrim is necessary. How to simultaneously optimize different r_{med} values for individual aerosol species?*

Reply: We added a brief description and reference to Lagarias, 1998 for the reader in the Method section:

> As an optimisation algorithm for the loss function $\mathcal{L}$ we use Nelder-Mead with free bounds, a popular minimisation method belonging to the class of direct search methods, i.e. that do not require knowledge of the gradients of the function, for functions of the kind $f(\mathbb{R}^n) \to \mathbb{R}$, which is the case for the constructed $\mathcal{L}$. The algorithm uses n-dimensional convex hulls and performs

> a series of reflection, expansion and contraction operations to converge on function minima (see e.g. Lagarias et al., 1998). Over the many setups used for the optimisation procedure, we never encountered important convergence issues.

14) *Page 12, section 4.3: please also discuss how the temporal co-location and averaging are applied.*

Reply: see above main question b) on the temporal sampling.

15) *Page 13, Table 3: Please discuss values in the 4th column (ClBase3).*

Reply: A separate Results subsection is now included:

> **5.3 N_d from below cloud-base aerosol (ClBase3)**
>
> If N_d errors. However, to our knowledge, GFAS wildfire emissions in the NTA and CAF regions are unlikely to suffer from so strong a positive bias able to explain a four- to five-fold error in simulated are diagnosed from aerosols located three model levels below cloud base, optimal PSDs provide a strongly reduced J for Sul ($\times 0.44$), Nit ($\times 0.52$) and Org ($\times 0.16$), and higher J for Ssf ($\times 2.55$) (see Table 4). For all species except Ssf, the optimal r_{med} are significantly larger than for the InCloud setup. For the optimised ClBase3 setup, $\mathcal{L} = 1.38 \pm 0.03$; this setup is markedly better than InCloud at matching MODIS N_d as illustrated in due to the lower $N_d^{1/3}$ RMSE (Figure 3) at latitudes higher than 30°S over the ocean, where $|\mathrm{RDND}| \lesssim 0.15$.

And discuss it in detail during the discussion:

> The optimal N_d values from ClBase3 optimisation perform markedly better than InCloud at mid- and high southern latitudes. This likely does not reflect better physical realism, because of the inconsistency with vertical tracer transport to cloud top (see

> discussion in Section 5), but rather a more meridionally-uniform aerosol background found below cloud base, than inside the cloud. To illustrate this point, Figure 14 shows the amount of diagnosed Ssf particles (represented with the prior PSDs) during the InCloud and ClBase3 setup. In principle, low Ssf number concentrations are expected, as sea-spray is secondary CCN provider (e.g. McFarquhar et al., 2021). However, coarse sea salt significantly impacts N_d in pristine regions via activation competition mechanisms of heterogeneous aerosol populations (Fossum et al., 2020), making a realistic representation of marine aerosol profiles essential to predict N_d. While particle concentrations typically between $\approx 7\,\mathrm{cm}^{-3}$ and $\approx 20\,\mathrm{cm}^{-3}$ are found at cloud level in subtropical latitudes, the concentrations sharply fall below $5\,\mathrm{cm}^{-3}$ with increasing latitude. Such meridional contrast is not found when diagnosing Ssf CCN just below the cloud base (with significantly higher values up

16) *Page 19, Figure 8: What is $N_{d,modis}$? Which of Q06, G18, and BR17?*

Reply: Thank you, that was unclear. It is the optimization target Q06, and now the figure and the caption make this explicit.

17) *Page 20, Figure 9: How is the IFS "ctrl" simulation configured? Would be useful to compare the simulations with original and modified r_{med} values.*
Reply: The now Figure 7 includes now also the simulated spectra using the optimised PSDs. We make the discussion also more detailed in the text, since the spectra integrated in the broader evaluation with additional hints that the optimal PSD for organic matter is unrealistic:

> and larger, eventually the optimal PSD returned for organic matter (OM) aerosol is inconsistent with independent AERONET observations over tropical Africa. This likely reflects limitations in the model representaton of black carbon ageing processes, that do not capture the clustering over time of soot particles into larger, internally-mixed aged aerosol. Possible future investigations could concern the accuracy of the described wildfire regime, as well as smoke plume properties, with downstream impact on carbonaceous aerosol vertical profiles and ageing timescales. To illustrate the diagnostic potential of ACI for constraining CAMS aerosol processes, we experimented two model versions that introduced modifications of the wet scavenging scheme. These target process representations that led to overestimated scavenged aerosol mass fraction in mixed-phase clouds. These changes improved the number mixing ratio of CCN inside mixed-phase clouds, as well as the simulated total-column aerosol particle numbers and AOD during the austral summer. This resulted in successful elimination of the top-of-the-atmosphere broadband shortwave bias over the Southern Ocean introduced by using the activation scheme with the CAMS aerosols in the current model version. CAMS aerosols can overall produce realistic CCN number concentrations and, in turn, aerosol. Excluding black carbon from the optimisation yields comparable N_d, without degrading aerosol optical properties.

Reviewer 2 - Comment 1

1. *The implementation of aerosol–cloud interactions in weather and climate models has been a longstanding topic of research. Numerous global climate models, seasonal forecasting systems, and modern reanalyses already include such representations (e.g., Seinfeld et al., 2016; Benedetti and Vitart, 2018; Wang et al., 2021; Song et al., 2025). Additionally, aerosol activation parameterizations have undergone extensive development over the past decades, resulting in computationally efficient schemes (e.g., Ghan et al., 2011). Retrievals of CCN concentrations from aerosol reanalyses such as CAMS have also been demonstrated (Block et al., 2024). A more thorough discussion of this existing body of work would have helped guide the authors' methodology and contextualize their contribution.*
Reply: We welcome the call of the reviewer for a more thorough discussion of the existing literature, and we thank them for highlighting the broader literature on ACI representation. We expanded the introduction to better contextualise our work within existing global models and activation parametrisations. We note that some of the cited studies have a different focus or cover aspects complementary to the aim of the introduction. Besides this, incorporating the relevant literature helped us better clarifying how our approach adds to existing work:

a single hygroscopicity parameter κ (Quaas and Gryspeerdt, 2022). Rothenberg and Wang (2016) used this framework in the implementation of an adiabatic parcel lifting model to accurately simulate the maximum supersaturation reached during the ascent of an air parcel . This allows the model to also Since the early formulations by Twomey in the late 1950s, activation models have been produced that can accurately solve the equations derived by the κ-Köhler theory for the air parcel lifting condensation problem (see Ghan et al. (2011) for a detailed review). In addition, climate models deployed fast physical parametrisations and lookup tables that have proven to perform satisfactorily compared to the full numerical models (Ghan et al., 2011).

Some numerical parametrisations can also represent activation of heterogenous aerosol mixtures and kinetic limitations of the activation process (Nenes et al., 2001; Ghan et al., 2011). These are needed to capture the effect of large sea salt particles to prevent on suppressing activation of available sulfate (SO_4^{2-}) aerosols (Fossum et al., 2020). It is, however, unclear what relevance this effect has for realistic aerosol populations and scenarios (Fossum et al., 2020). The relevance of this effect

and

medium-range (up to 10 days) and longer timescales (months). A major goal of this study was to associate recent study by Block et al. (2024) showed that the aerosols from the CAMS reanalysis (CAMSRA) can provide CCN diagnostics for validation against in-situ observations, but also revealed systematic errors hinting at underlying model issues. This illustrates the need for improved model aerosol descriptions to induce as realistic as possible values of droplet number concentrations (global CCN concentrations and N_d) values in liquid cloudsto aerosol mass mixing ratios (MMR) in the model. In order to , which

constitutes the main goal of this study. To achieve this, we designed a method to efficiently constrain simulated number concentrations from prognostic aerosol using an 18-years long 18-year dataset of MODIS N_d retrievals . spanning 2003–2020.

References provided by Reviewer 2 for this question:

- Benedetti, A., & Vitart, F. (2018). Can the direct effect of aerosols improve subseasonal predictability? *Monthly Weather Review*, 146, 3481–3498. https://doi.org/10.1175/MWR-D-17-0282.1
- Block, K., Haghighatnasab, M., Partridge, D. G., Stier, P., & Quaas, J. (2024). Cloud condensation nuclei concentrations derived from the CAMS reanalysis. *Earth System Science Data*, 16, 443–470. https://doi.org/10.5194/essd-16-443-2024
- Ghan, S. J., Abdul-Razzak, H., Nenes, A., Ming, Y., Liu, X., Ovchinnikov, M., Shipway, B., Meskhidze, N., Xu, J., & Shi, X. (2011). Droplet nucleation: Physically-based parameterizations and comparative evaluation. *Journal of Advances in Modeling Earth Systems*, 3(4). https://doi.org/10.1029/2011MS000074
- Seinfeld, J. H., Bretherton, C., Carslaw, K. S., et al. (2016). Improving our fundamental understanding of the role of aerosol–cloud interactions in the climate system. *Proceedings of the National Academy of Sciences*, 113(21), 5781–5790. https://doi.org/10.1073/pnas.1514043113
- Song, C., McCoy, D., Molod, A., Aerenson, T., & Barahona, D. (2025). Signatures of aerosol–cloud interactions in GiOcean: A coupled global reanalysis with two-moment cloud microphysics. *Atmospheric Chemistry and Physics*, 25, 15567–15592. https://doi.org/10.5194/acp-25-15567-2025
- Wang, C., Soden, B. J., Yang, W., & Vecchi, G. A. (2021). Compensation between cloud feedback and aerosol–cloud interaction in CMIP6 models. *Geophysical Research Letters*, 48(4), e2020GL091024. https://doi.org/10.1029/2020GL091024

2. *The manuscript does not clearly define what is meant by the implementation of ACIs. Most of the discussion focuses on adjustments to the aerosol size distribution and the resulting changes in aerosol fields, with minimal reference to cloud properties. It remains unclear whether the computed Nd is actively used to influence or update any cloud-related processes in the model, raising doubts about the actual implementation of ACIs.*

   Reply: This point is similar to Reviewer 1 Comment 1, and we refer to the reply above. In synthesis, we extended the manuscript by including a description of the cloud model and explicitly stated in the abstract, introduction, method and conclusions that the ACI are intended within the study up to the first indirect radiative effect, providing contextual information and the rationale behind this choice.

3. *The assumption of a constant updraft velocity of 1 m/s lacks physical justification and is not supported by observational or theoretical evidence. It is well established that Nd is highly sensitive to updraft velocity, which has profound implications for the aerosol indirect effect (Sullivan et al., 2016). While it is true that updraft is among the most uncertain parameters in ACI modeling, applying a fixed and globally unrealistic value, appropriate only for small marine cumulus clouds, is not defensible.*

   Reply: We agree and have substantially improved the treatment of vertical velocities for activation, as will be described in the updated version of the manuscript. Firstly, the resolution of the tuning procedure has been brought from 3x3 degrees to 1x1 degrees for a sharper identification of cloud regimes (and finer than many climate models). We now include the large-scale vertical wind as mean vertical velocity (as in Van Noije, 2021) and the Deardorff convective scale velocity w* (as defined in Deardorff, 1970). Many models determine vertical velocity scales w' from turbulent kinetic energy (TKE), which is however not viable for us during the offline tuning procedure due to the limited number of diagnostics available in the ERA5 datasets, and no TKE. While using w* as a diagnostic for TKE would miss non-convectively induced turbulence, especially related to cloud top radiative cooling, a satisfactory characterization of vertical updraughts for aerosol activation is still a challenge for global models and these typically need to resort to setting minimum boundaries for the vertical velocity standard deviation σ_w (e.g. 0.1 m/s in Barahona, 2014 and 0.7 m/s in Golaz, 2011), that are reported by Golaz, 2011 to occur more than 90% of the time, and reliable parametrizations for updraft velocities in marine boundary layer clouds are an open area of research (see e.g. Ahola, 2022 and references therein). With these changes, the activation scheme reaches a level of sophistication comparable to many in the literature:

and Wang, 2016). We perform simulations for a range of vertical velocities w (updrafts) to precisely describe also regimes with relatively high CCN concentrations (greater than $1000\ \mathrm{cm}^{-3}$- so called updraught-limited) (Pruppacher and Klett, 2010;

phase regimes (above $0\ \mathrm{K}$) (Leaitch et al., 1986; Rothenberg and Wang, 2016). We perform simulations for a range of vertical velocities w (updrafts) to precisely describe also regimes with relatively high CCN concentrations (greater than $1000\ \mathrm{cm}^{-3}$- so called updraught-limited) (Pruppacher and Klett, 2010; Reutter et al., 2009; Rothenberg and Wang, 2016; West et al., 2014). However, diagnosing accurate sub-grid vertical velocity and variability in a hydrostatic model with a purely-diagnostic description of turbulence can be quite a challenge; this also poses the risk of introducing significant noise in the results, rendering the identification of major sources of error, namely aerosol number concentrations, more difficult in such a first implementation of the activation scheme. Therefore, the simulations are done for every combination of number concentration for each species over 7 geometrically-spaced values, using a fixed vertical velocity of $1\ \mathrm{m/s}$ as representative for boundary layer mixing velocities (Hogan et al., 2009), resulting in 117649 simulations. , so called updraft-limited) (Pruppacher and Klett, 2010; Reutter et al., velocity of 1 $\mathrm{m/s}$ as representative for boundary layer mixing velocities (Hogan et al., 2009), resulting in 117649 simulations. In order to , updraft-limited) (Pruppacher and Klett, 2010; Reutter et al., 2009; Rothenberg and Wang, 2016; West et al., 2014; Sullivan et al., 2016). To keep the number of simulations lowmanageable, we define a small set of 6 internally-mixed CCN CCN-providing aerosol species derived from aerosol species defined those in Table 1, by grouping together those with similar size and species with similar physico-chemical properties and assigning hygroscopicity κ reflecting typical values with typical values inferred from Petters and Kreidenweis (2007). The CCN species used for the offline activation simulations, as well as the definitions mapping the IFS aerosols into the CCN species (recipes) are listed Their definitions are reported in Table 2. Results of the activation process are stored in a lookup table (LUT) as critical dry activation radii,as well as corresponding activated number and mass fractions. We performed activation simulations for every combination of number concentration for each species and vertical velocity (between $0.1\ \mathrm{m\,s^{-1}}$ and $10\ \mathrm{m\,s^{-1}}$) over seven geometrically-spaced values resulting in a total of 823,543 simulations. Subsequently, these results were integrated over Gaussian distributions of vertical velocities with seven values of mean $\bar{w}$ in the range $[-0.2\ \mathrm{m\,s^{-1}}, 0.2\ \mathrm{m\,s^{-1}}]$ (symmetric around zero, absolute values geometrically spaced) and seven geometrically-spaced values of standard deviation σ_w in the range $[0.05\ \mathrm{m\,s^{-1}}, 2.5\ \mathrm{m\,s^{-1}}]$. The integration is performed only for $w > 0$ as in Golaz et al. (2011). The final size of the LUT is of 5,764,801 elements.

The LUT yields activated number fraction (a_x) for each CCN-providingspecies x listed in table 2 given a Table 2 for a given mixture of particles . Such values are used to compute the amount of aerosol particles activated into droplets for each species. The and configuration of $\bar{w}$ and σ_w. The activation scheme is in this sense a wrapper of the LUT accessor with pre- and post-processing capabilities, that extracts the nearest-neighbor value along the aerosol concentrations dimensions, and linearly interpolates along the $\bar{w}$ and σ_w dimensions. As a final step, the scheme computes the number of droplets N_d is defined as the

and in the scheme implementation

In the setup for this study, the IFS radiation scheme can diagnose N_d with a call to the activation scheme described above by providing as input $\bar{w}$, σ_w and aerosol mass concentrations of all aerosol species required to construct the CCN providers of Table 2. $\bar{w}$ is set equal to the large-scale vertical velocity, which is typical for climate models (Ghan et al., 2011; Van Noije et al., 2021). σ_w for turbulent boundary layers is generally diagnosed from turbulent kinetic energy (TKE) or eddy diffusivity (Ghan et al., 2011); however, these are not available as ERA5 fields, therefore for this study we used the Deardorff velocity scale w^* for convective boundary layers (Deardorff, 1970), scaled by a factor 0.6 to diagnose σ_w, in line with relationships found in observations and numerical simulations (Deardorff, 1970; Hogan et al., 2009). A minimum value of $\sigma_w = 0.1\ \mathrm{ms^{-1}}$ is imposed to ensure physical activation conditions (e.g. Golaz et al., 2011; Barahona et al., 2014).

4. *The treatment of satellite retrieval uncertainties is problematic. The authors appear to treat differences arising from alternate retrieval assumptions as equivalent to experimental error, which is not technically sound. Moreover, model outputs must be sampled in a manner consistent with the assumptions of the satellite retrievals to avoid artificial biases. For instance, retrieval filters based on temperature and cloud fraction tend to exclude clouds with low Nd, particularly in high-latitude regions. If such filters are not consistently applied to the model data, it can lead to systematic biases, potentially causing the authors to erroneously tune scavenging parameters in response to what is essentially a sampling artefact.*

Reply: Satellite retrievals of Nd from spectrometers are notably affected by substantial uncertainties, largely depending on the cloud-adiabaticity assumption and the accuracy of LWP / r_{eff} (Gryspeerdt, 2022). We would like to clarify that the datasets produced by Gryspeerdt, 2022 used different sampling strategies rather than different retrieval assumptions, within an approach explicitly aimed at reducing uncertainties “through sampling retrievals with higher confidence” (Gryspeerdt, 2022). While agreeing that this does not imply a precise quantification of the uncertainty of these retrievals, our methodology relies on implicitly assuming that the spread across these datasets is a proxy quantity for the uncertainty of the retrieval, and we assume that it is proportional to it in order to construct a well-defined weighting factor for the loss function. Despite the simplification implied by our approach, this represents a step forward with respect to previous relevant studies using MODIS Nd data to constrain model representation (see e.g. McCoy, 2018), and explicitly acknowledge in the manuscript that this highlights the need for improved global-scale quantifications of Nd retrieval uncertainties. About the concerns on the sampling of model data, we indeed applied filtering on cloud-top temperature, phase, and total optical thickness as done with the satellite retrievals, to avoid introduction of sampling biases for the comparison as reported in section 4.2 (Model data selection). Due to the much lower resolution of the model data, it was necessary to weaken the filtering criteria when applying them to gridbox-mean quantities. As detailed in the reply to Reviewer 1 Comment 3, upgrading the entire system to 1x1 degrees data brings the model data selection criteria significantly closer to those of the observations.

Finally, concerning the reviewer's remark that such sampling issues eventually could lead to systematic biases to be corrected by changing the wet scavenging parametrization, we would like to clarify that the study does not involve any tuning of the scavenging parameters to improve the comparison between simulated and satellite N_d. The motivation for investigating the wet scavenging parametrization instead arises from the results of the InCloud setup and the Figure 8 (in the updated manuscript) model cross-section, showing virtually no aerosols inside the clouds over the Southern Ocean. The new structure of the manuscript, with a clear separation between the results of the optimisation and the wet scavenging sensitivity experiments should help make the text clearer on this point:

6 Testing changes in the aerosol wet-scavenging scheme

In order to investigate the low CCNconcentrations at cloud level we The results presented in Section 5 reveal a pattern of low biases at mid- to high latitudes (above 60°N and 45°S), that persist at the end of the optimisation. In these regions, aerosol concentrations tend to decrease sharply in the in-cloud relative to the below-cloud environment, typically coinciding with precipitating mixed-phase clouds. This motivated some targeted sensitivity tests with online experiments to explore the relevance of the wet removal of aerosols on the availability of simulated CCN.

We initially collected diagnostics of in-cloud (rain-out, IN-SCAV) and below-cloud (wash-out, BC-SCAV) wet scavenging (SC) as well as the release by precipitation evaporation (EVAP). These confirmed our expectation that IN-SC confirm the expectation that IN-SCAV from large-scale precipitation dominates the removal of SU and SS, while EVAPis significant in

5. *The global optimization approach based on satellite retrievals fails to account for their limited validity. Retrieval techniques are typically applicable to vertically homogeneous, adiabatic, low-level clouds, which are the exception rather than the norm. Despite this, the authors extend the use of the retrieval to mixed-phase clouds and regimes strongly influenced by convection, where the assumptions underlying the retrieval no longer hold.*
Reply: The selection of model data described in section 4.2 is aimed at filtering out clouds in convective-dominated regimes. Thanks to the reviewer's remark, we realized that we inadvertently omitted to specify that we adopted a cloud cover criterion (at least 0.8 on 3x3 degrees grid), in addition to the described

criteria on cloud top temperature (>283K) and the presence of cloud-top ice water content (must be less than 50% of total water content on 3x3 degree grid). The combination of these makes it unlikely for clouds strongly dominated by convection to be selected for the optimization procedure. As correctly pointed out by the reviewer, this was a critical aspect of the study, and in the updated version of the paper we use 1x1 degrees data and tighten the ice water content criterion (<0.1 of total water) to make the selection of cloud model data more consistent with the satellite retrievals. Details are given in the response to similar Reviewer 1 Comment 3.

6. *The manuscript does not include a data availability statement. The code availability statement does not refer to the parcel model, nor the lookup table used in this work. Given the nature of the work and its reliance on numerical model development and evaluation, it is essential that both the data and the implementation code be made publicly accessible to support reproducibility and validation.*

   Reply: The parcel model is referenced by citing Rothenberg and Wang (2016) as indicated in the model's documentation (https://pyrcel.readthedocs.io/en/latest/). We now provide a DOI for the lookup table along with a reference to the offline optimization code:

   > *Code and data availability.* The IFS code is the intellectual property of ECMWF and its member states, and therefore version CY49R1 used for this study is not yet publicly available. However, access to the IFS code, including IFS-COMPO, updated to version CY48R1 is provided by the OpenIFS project (https://github.com/ecmwf-ifs/openifs). The code used to simulate optical properties uses Scott Prahl's Miepython library (https://github.com/scottprahl/miepython). The lookup table for activation used for this study can be accessed at https://doi.org/10.5281/zenodo.20051869. The code used for the offline optimisation is available at https://github.com/pandreozzz/optimind.

References

Ahola, Jaakko et al. (2022). "Technical Note: Parameterising Cloud Base Updraft Velocity of Marine Stratocumuli". In: *Atmospheric Chemistry and Physics* 22.7, pp. 4523–4537. doi: 10.5194/acp-22-4523-2022.

Barahona, D. et al. (2014). "Development of Two-Moment Cloud Microphysics for Liquid and Ice within the NASA Goddard Earth Observing System Model (GEOS-5)". In*: Geoscientific Model Development* 7.4, pp. 1733–1766. doi: 10.5194/gmd-7-1733-2014.

Deardorff, James W. (1970). "Convective Velocity and Temperature Scales for the Unstable Planetary Boundary Layer and for Rayleigh Convection". In: *Journal of the Atmospheric Sciences* 27.8, pp. 1211–1213. doi: 10.1175/1520-469(1970)027<1211:CVATSF>2.0.CO;2.

Golaz, Jean-Christophe et al. (2011). "Sensitivity of the Aerosol Indirect Effect to Subgrid Variability in the Cloud Parameterization of the GFDL Atmosphere General Circulation Model AM3". In: *Journal of Climate* 24.13, pp. 3145–3160. doi: 10.1175/2010JCLI3945.1.

Grosvenor, Daniel P., Odran Sourdeval, and Robert Wood (2018). "Parameterizing Cloud Top Effective Radii from Satellite Retrieved Values, Accounting for Vertical Photon Transport: Quantification and Correction of the Resulting Bias in Droplet Concentration and Liquid Water Path Retrievals". In: *Atmospheric Measurement Techniques* 11.7, pp. 4273–4289. doi: 10.5194/amt-11-4273-2018.

Gryspeerdt, Edward et al. (2022). “The Impact of Sampling Strategy on the Cloud Droplet Number Concentration Estimated from Satellite Data”. In: *Atmospheric Measurement Techniques* 15.12, pp. 3875–3892. doi: 10.5194/amt-15-3875-2022.

Jones, Andy et al. (2001). “Indirect Sulphate Aerosol Forcing in a Climate Model with an Interactive Sulphur Cycle”. In: *Journal of Geophysical Research: Atmospheres* 106.D17, pp. 20293–20310. doi: 10.1029/2000JD000089.

Lagarias, Jeffrey C. et al. (1998). “Convergence Properties of the Nelder–Mead Simplex Method in Low Dimensions”. In: *SIAM Journal on Optimization* 9.1, pp. 112–147. doi: 10.1137/S1052623496303470.

Leaitch, W. R. et al. (1986). “Cloud Droplet Nucleation and Cloud Scavenging of Aerosol Sulphate in Polluted Atmospheres”. In: *Tellus B* 38B.5, pp. 328–344. doi: 10.1111/j.1600-0889.1986.tb00258.x.

Mitchell, D. L.: Parameterization of the Mie Extinction and Absorption Coefficients for Water Clouds, Journal of the Atmospheric Sciences,
57, 1311–1326, https://doi.org/10.1175/1520-0469(2000)057<1311:POTMEA>2.0.CO;2, 2000.

McCoy, Daniel T. et al. (2018). “Predicting Decadal Trends in Cloud Droplet Number Concentration Using Reanalysis and Satellite Data”. In: *Atmospheric Chemistry and Physics* 18.3, pp. 2035–2047. doi: 10 . 5194 / acp -18-2035-2018.

Rémy, Samuel et al. (2022). “Description and Evaluation of the Tropospheric Aerosol Scheme in the Integrated Forecasting System (IFS-AER, Cycle 47R1) of ECMWF”. In: *Geoscientific Model Development* 15.12, pp. 4881–4912. doi: 10.5194/gmd-15-4881-2022.

Rothenberg, Daniel and Chien Wang (2016). “Metamodeling of Droplet Activation for Global Climate Models”. In: *Journal of the Atmospheric Sciences* 73.3, pp. 1255–1272. doi: 10.1175/JAS-D-15-0223.1.

Van Noije, Twan et al. (2021). “EC-Earth3-AerChem: A Global Climate Model with Interactive Aerosols and Atmospheric Chemistry Participating in CMIP6”. In: *Geoscientific Model Development* 14.9, pp. 5637–5668. doi: 10.5194/
gmd-14-5637-2021.

Wang, Chunpeng et al. (2014). “A Physically Based Algorithm for Non-Blackbody Correction of Cloud-Top Temperature and Application to Convection Study”. In: *Journal of Applied Meteorology and Climatology* 53.7, pp. 1844–1857. doi: 10.1175/JAMC-D-13-0331.1.

West, R. E. L. et al. (2014). “The Importance of Vertical Velocity Variability for Estimates of the Indirect Aerosol Effects”. In: *Atmospheric Chemistry and Physics* 14.12, pp. 6369–6393. doi: 10.5194/acp-14-6369-2014