Reply to the comments for egusphere-2025-132

We thank the reviewer and the editor for the additional comments which further helped us improved the manuscript. We have provided replies to the comments and revisions in the manuscript. The reviewer's comments are shown below in black, and our responses are provided in blue. New sentences and paragraphs have been added to the manuscript, highlighted in green italics. Line numbers refer to those in the revised manuscript.

- L11: "However, as it becomes closer to the source…" This sentence is unclear; please consider rephrasing for clarity.

Reply: Line 11 has been revised as follows: "*However, nucleation is suppressed very close to the pollution source, resulting in lower nucleation and soluble Aitken mode aerosol number concentrations*."

- L14: Cloud water mass and rain mass are not standard terms to my knowledge. Should it be mass mixing ratios?

Reply: Yes, they are mass mixing ratios and have been corrected in the abstract.

- L16: This may be a detail, but it seems here that a weak response is taken to imply non-linearity here. A response can be weak but linear. It is only non-linear if the response changes depending on the perturbation strength. It is not clear to me in the context what the non-linearity refers to here.

Reply: We agree that a weak response does not necessarily relate to non-linearity, and we will specify the two findings explicitly. The response of cloud and rain mass mixing ratios is weak when averaged over cloudy and or high-sulfur regions and does not scale linearly with the changes in anthropogenic emissions across the simulations with scaled anthropogenic emissions. When we altered the number of aerosols in the cloud microphysics scheme by factors of 0.25 and 4, the corresponding changes in cloud droplet number concentration was large (by around factors of 0.5 and 2). However, such significant changes in droplet number still resulted in moderate changes in cloud mass mixing ratio and precipitation. It indicates that the pathway from aerosol perturbations to rain formation is buffered by several microphysical processes and the overall response is non-linear.

The sentence at line 16 has been revised to "*If we assume our simulation has a fine enough grid resolution and an accurate representation of the relevant atmospheric processes, the simulated weak and non-linear response of cloud and rain properties to linearly scaled anthropogenic emissions suggests that the interactions among aerosol, cloud and precipitation in the Amazonian convective environment are buffered by microphysical processes*."

- Fig. A11. Is it really so that the ratio of Aitken mode to insoluble Aitken mode is on the scale of 0 to 1e10? This seems a bit like it could be a unit error. Also, is it Aitken soluble over Aitken insoluble? Or Aitken total over Aitken insoluble? I suggest to make this clear in the caption.

R: It is soluble Aitken mode vs insoluble Aitken mode concentrations around 550 m altitude and have been corrected in the caption.

The ratio of soluble Aitken mode to insoluble Aitken mode number concentrations is unitless. The concentrations are collected around 550 m altitude where NPF is permitted, while this altitude is much higher than pollution source (at the surface). Thus, insoluble Aitken mode aerosol concentrations are low compared to soluble Aitken mode aerosol at this altitude, resulting in high ratios. We checked the distributions of the ratios and found that the majority of the ratios are between 10^2-10^5. Those reaching up to 10^{10} are rare (see the figure below).

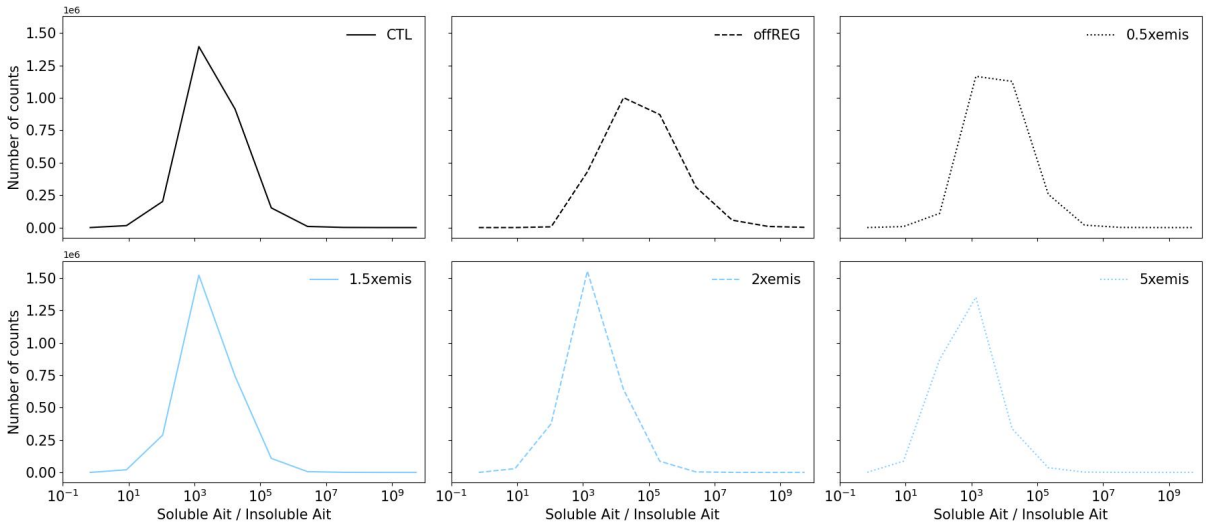

Fig. Histograms of the ratios of soluble Aitken mode to insoluble Aitken mode aerosol number concentrations at 555 m altitude.

- L264: About the description of the scaling in the 0.25xaero and 4xaero simulations: I found your response to the review comment much clearer than the current manuscript text. Consider incorporating something like: "This means we directly scale the number of particles after the activation diameter is determined, and thus do not allow the scaling to influence supersaturation, activation diameter, etc."

R: We revised the sentence to "*In this procedure, we directly scale the number of particles after the maximum supersaturation has been determined and thus do not allow the aerosol activation diameters and concentrations to be adjusted to updraft velocities or water vapour availability*." The sentence is now at line 266.

- L513-518: This sentence should be split for clarity. I also don't understand what "but we expect the absence of isoprene-NOx mechanism to be within the uncertainty of current NPF mechanism" means.

R: Yes, we agree that the sentence is better when split. We have moved the content regarding isoprene-NO_x nucleation to the end of the paragraph.

We expect the effects of the absence of isoprene-NO_x mechanism to be within the uncertainty of the current NPF mechanism, especially given that we have to disable upper tropospheric NPF to better match the observed particle concentrations. To better match the observations, we have investigated a range of oxidation rates of monoterpene to form HOMs to particle concentrations in the Amazon during the wet season and the dry season (Wang et al., 2023) which is to some extent will produce similar variability to the addition of a NPF mechanism based on isoprene-NO_x. The results indicated that enabling NPF in the upper troposphere could always produce around 1000 cm^{-3} particles in the free troposphere which is much more than those observed during the GoAmazon2014/5 campaign.

We deleted the sentence to avoid confusion and added "*Our model does not include this NPF mechanism due to the absence of isoprene-NO_x chemistry, but we do not expect the absence of this mechanism to significantly affect our results.*" at line 518.

- L518-520: I am not sure I understand what your conclusion is for point 2) here. Are you saying that N100 may be wrong due to upper tropospheric nucleation, or that you think this is unlikely?

R: Incorporating upper tropospheric (UT) NPF causes the model to significantly overestimate the observations. It does not necessarily mean that including UT NPF is wrong, but rather indicates a mismatch between the model and the observations. Not using UT NPF is not physically realistic because many previous studies have shown the importance of new particle formation (NPF) in the UT. Therefore, UT NPF is generally included in the models. In this study we only use the GoAmazon2014/5 observation dataset for our time period, which focused mainly on boundary layer and found very few particles in the free troposphere during the Amazonian wet season.

When we switched on UT NPF in the model, particle concentrations increased significantly in the free troposphere and the boundary layer, leading to an overestimation compared to the observations. To better match the observations, we have to suppress UTNPF in our simulations. This setup is not ideal but a compromise that likely still causes biases in the concentrations of particles greater than 100nm in diameter, and should be improved on in future simulations..

It is important to note that the aircraft measurements were primarily focused on the boundary layer and it is very uncertain how representative the observations in the free troposphere are of typical conditions in that region.

This paragraph starting from line 514 has been revised as follows:

"*(2) Upper tropospheric (UT) NPF along with subsequent downward transport, has been shown to be important for determining low-level particle concentrations (Clarke et al., 1998, 1999; Clarke and Kapustin, 2002; Merikanto et al., 2009; Wang et al., 2016; Williamson et al., 2019; Curtius et al., 2024), and it is important for Amazonia during the dry season (Andreae et al., 2018). Observations have reported bursts of particles due to NPF from organic compounds formed by isoprene with NO_x (Kuhn et al., 2010; Bardakov et al., 2024; Shen et al., 2024). Our model does not include this NPF mechanism due to the absence of isoprene-NO_x chemistry, but we do not expect the absence of this mechanism to significantly affect our results. In this study we focus on the wet season and only use the GoAmazon2014/5 observation dataset for our time period, which focused mainly on the boundary layer (below 2 km), although the infrequent sampling casts doubt on how representative of mean conditions these observations were. The aircraft occasionally flew between 2 - 6 km altitude and found very few particles in the free troposphere during the Amazonian wet season. Consequently, it is very uncertain how representative the observations in the free troposphere are of typical conditions in that region. When we switched on UT NPF in our model, particle concentrations increased significantly in the free troposphere and the boundary layer, leading to an overestimation compared to the observations. To better match the observations, we therefore disabled NPF above 1 km to achieve consistency between the model and observations in March 2014. This setup is not ideal but a compromise that likely still causes biases in the concentrations of particles greater than 100nm in diameter, and should be improved on in future simulations*."

- L571: I thought I had understood what you had done with the 0.25 and 4xaero simulations, but it is not clear to me why you are then getting a reduction in Nd which is half of your perturbation? Could you clarify?

R: The 'N_i' that we perturbed in the activation scheme is not equivalent to the N_d in the model output. Although the variable 'N_i' is scaled by a factor of 0.25 and 4, the N_d in the model output is influenced by several nonlinear microphysical (to aerosol number concentrations) processes every timestep (the Abdul-Razaak and Ghan (2000) droplet activation scheme itself, rain evaporation, formation of graupel, deposition, condensation etc.). Additionally, N_d in Fig. 7 is derived after averaging the 3-hourly output over time and cloudy-polluted regions. As a result, the changes in N_d relative to CTL simulation are not linear to the aerosol perturbations (0.25 and 4). We added the following at line 581 to clarify it:

"*The variable we perturbed in these simulations (N_i in the Abdul-Razaak and Ghan (2000) droplet activation scheme, hereafter ARG2000) in the CASIM activation scheme is not directly equivalent to the model output N_d since ARG2000 is non-linear between N_i and N_d. The model output N_d depends strongly on updraft speeds via the activation parameterization, and is also influenced several dynamical and microphysical processes each timestep (e.g. advection, droplet freezing, riming, or warm rain formation) and N_d shown in Fig. 7 has been averaged over time and high-sulfur-cloudy regions using the 3-hourly model output. Therefore, changes in N_d in these simulations do not scale directly with the aerosol perturbation relative to the CTL simulation*."