

## Reply to Anonymous Referee #2

We are grateful to the referee for her/his useful suggestions to our manuscript. Please find below a detailed point-by-point reply (referee's comment in *italic*).

*p.5, lines 3-4. "To avoid interpretation of. . ." is a bit confusing. I would suggest "To avoid convoluting the results with feedbacks from. . ."*

Thank you for this suggestion, which we have implemented.

*p.5, line 7. If the "big leaf" approach is mentioned here, a reference should be provided.*

We have added a reference to Sellers et al. (1996).

*p.6, lines 12–20. This paragraph is a little confusing. The criterion for transfer to mixed modes is stated as "a liquid coating of a critical size" (size presumably meaning thickness here?), but with aerosol water neglected. Surely the thickness of a liquid coating would be largely determined by the amount of water, however? Or does this rather mean a soluble coating of a critical thickness, e.g. a layer of sulphate  $n$  molecules thick, ignoring any water? Such a coating-thickness criterion would require an amount of soluble material proportional to the surface area of the insoluble core, while the following text describes a criterion as a proportion of the mass or volume. Please clarify.*

We have rephrased this paragraph as follows: "In the original version of the model, the transfer to the mixed modes was induced as soon as insoluble particles obtained a liquid coating of a critical size. We now neglect aerosol water in this aging calculation and, correspondingly, in the target mode assignment upon particle coagulation. Hence only the water-soluble components of the coating are taken into account. In this way we interpret water uptake as a consequence of particle aging rather than as cause of it. We further neglect the POM fraction in these model operations since its role in the aging process is still uncertain. Particles from the insoluble modes are now transferred to the mixed modes if the sum of the soluble inorganic component masses exceeds 10 % of the modal dry mass. This assumption is supported by laboratory and field measurements as reported by Svenningsson et al. (1994), Khalizov et al. (2009), and Liu et al. (2013). Correspondingly, we assign particles that result from coagulation of insoluble modes with mixed or soluble modes to an insoluble mode when the resulting soluble inorganic contribution to dry mass is less than 10 %, and to a mixed mode otherwise."

*p.6, line 21. Please explain the rescaling which is applied here.*

We have added the following sentence to clarify: "This basically corresponds to redistributing the nucleated mass into larger particles, assuming a lognormal size distribution with the same width but with a larger median diameter, which results in a decrease in nucleated particle number."

*p.7, lines 3–5. This sentence seems to suggest the Bergeron-Findeisen process represents the freezing of cloud droplets, with the evaporation of droplets as an alternative. I assume this is a mistake in the phrasing, since (as the authors are no doubt aware) the Bergeron-Findeisen process represents the latter (evaporation of droplets, releasing their aerosol, with deposition of the resulting vapour onto existing ice particles) rather than the former (freezing of existing droplets).*

*Please clarify the sentence.*

We have rephrased the sentence accordingly: "This rough estimate is based on the fact that, due to the limited number of ice nuclei, only a fraction of cloud droplets freezes during glaciation of liquid clouds, while the majority of the droplets evaporates via the Bergeron-Findeisen process, thereby releasing large amounts of aerosol mass originally scavenged during liquid droplet formation."

*p.8, line 20. What about impaction by cloud droplets (not by precipitation, which is described separately)? Both activation and impaction are mentioned earlier as separate processes by which aerosol particles can be taken up by cloud droplets, but it is unclear whether insoluble particles are subject to impaction only, or to neither process, in the model.*

The model takes into account Brownian motion scavenging of insoluble particles within clouds. This is described in detail in Appendix B1. We have included a corresponding cross reference in Section 2.3 in order to enhance the readability.

*p.12, Table 3. There are a lot of dense numbers in this table, which would be clearer visualised in chart form.*

We have chosen the table form as this allows direct comparison with similar tables presented in other relevant papers, such as Righi et al. (2013), about a previous MADE version, and Pozzer et al. (2012). A visual summary of these results is already provided by Figure 2 and Figure 3.

*p.14, lines 3–4. This seems to put quite a positive spin on what looks from the figure like a general disagreement between model and observations. I would re-phrase this to more clearly acknowledge the large overall error, while still pointing out the agreement in the spatial pattern.*

Following a similar suggestion by Referee #1, we have rephrased this sentence as follows: “The model mostly reproduces the spatial pattern in this region, but it does not capture the west-east gradient seen in the observations, and is biased high.”

*p.22, lines 2–3. Please clarify which phase of the AeroCom project is referred to here, as the models have evolved significantly since Phase I.*

Good point. The cited study (Schwarz et al., 2013) used the Phase II models. We have added this.

*p.27, Figure 9. Do these observations really show more than 50% of the aerosol in the 46–167nm range being dust? That’s a surprisingly large fraction at such small sizes, and if correct represents the major disagreement with the model which should be discussed in the text.*

The large dust fraction could be a consequence of the specific location of the field campaign. Since Cape Verde is frequently impacted by the dust outflow of the Sahara (which was the major subject of the campaign) even high amounts of fine dust particles could have occurred. However, the measurements might have suffered from uncertainties particularly in this size range. We have extended the discussion of this issue in the manuscript (last paragraph of section 3.4): “Model misrepresentation, for instance, of the mineral dust particle size distribution, of the local sulfate concentration, or of the competition between nucleation and condensation of gaseous H<sub>2</sub>SO<sub>4</sub>, could also play a role. On the other hand, the SEM analysis in particular of the smallest size fraction might have a bias towards an underestimation of sulfate particles due to their instability under the electron beam. Since the number concentration of particles in this size fraction is comparatively high, a thorough analysis, including also comparisons of the measured and simulated size distributions and also measurement uncertainties, should be the subject of a separate study.”

*p.31, Table 5. There are a lot of dense numbers in this table, which would be clearer visualised in chart form.*

This is a very good suggestion. We now present the results in terms of bar charts (Fig. 11) rather than a table.

*p.3, line 31. Delete commas in “both, gases and aerosol particles, . . . ”*

*p.11, line 6. “of several 10%” doesn’t read well. I’d suggest “of several tens of percent”.*

*p.11, line 18. CASTNET and IMPROVE are mentioned here, but the acronyms are not defined until their next use on lines 31–33.*

*p.11, line 26. Delete comma after “both”.*

*p.16, lines 15–16. Change “statistics of. . . is” to “statistics of. . . are”.*

*p.17, line 29. Delete commas in “both, the simulation, and. . . ”*

*p.22, line 5. Delete commas in “both, spatial and temporal coverage, . . . ”*

*p.24, lines 24–25. “up to several 10%” doesn’t read well. I would suggest “up to several tens of percent”.*

*p.28, line 22. “lead to several 10% different. . . ” doesn’t read well. I would suggest “lead to several tens of percent difference in. . . ”*

*p.30, line 4. Delete “shows” in “. . . algorithm shows significantly overestimates AOD. . . ”*

*p.31, line 13. “several ten percent” doesn’t read well. I would suggest “several tens of percent”.*

All done. Thanks for spotting these typos.

*p.32, line 7. Change “size distributions, . . . , was. . . ” to “size distributions, . . . , were. . . ”*

The verb here refers to “level of agreement”, hence the singular form is correct.