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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol size distribution and radiative forcing response to anthropogenically driven historical changes in biogenic secondary organic aerosol formation" *by* S. D. D'Andrea et al.

Anonymous Referee #2

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The manuscript presents a model estimate of the aerosol-mediated climate impacts of millennial scale changes in biogenic volatile organic compound (BVOC) emissions. Overall, this is timely study that raises several important points (e.g., human impact on natural emissions, need to define the preindustrial state of the atmosphere robustly). The authors have performed a relatively comprehensive set of simulations to test the sensitivity of their results and, for the most part, the results are presented clearly. I was also very pleased to see that the authors openly discussed their model limitations and acknowledged many of the remaining scientific uncertainties that can impact their

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calculations. Therefore, I recommend the manuscript to be published in ACP after the following comments have been addressed.

My main criticism is that at points the discussion of the numerous simulations is confusing or even slightly misleading for the reader. Especially:

a. In section 4.3 the authors use the standard approach to calculate aerosol radiative effects, i.e. change from the "unperturbed" atmosphere (in this study simulation BE1.AE0) – this is all fine and makes comparison to other studies straightforward. However, based on the abstract (lines 16-21), I was for a long time under the impression that the radiative effects are calculated from the simulation with *present-day* anthropogenic emissions (which would be a confusing choice), and that the sensitivity simulations discussed directly underneath are built on this present-day emission scenario. (What adds to the confusion is that the regional effect >0.5 W/m2 from this scenario (line 21) is never even discussed in section 3.4.).

Overall, I am not convinced that the simulations with present-day anthropogenic emissions (which most of the sensitivity runs are) are very useful for the radiative effect calculations. Basically these simulations tell what the radiative effect have would be if the anthropogenic emissions had been at present-day level already in year 1000 – this is not a very realistic scenario. I therefore recommend that for the radiative effect calculations only the anthropogenic off simulations (AE0) should be presented. This will still allow discussion of the impact of BVOC emission and SOA yield uncertainty.

b. Furthermore, I do not agree that comparing simulations with and without present-day anthropogenic emissions (AE2 and AE0) would account for uncertainties in anthropogenic emissions (e.g., p. 26325, lines 4-5). AE0 for year 1000 is likely to be a fairly good assumption; however, for the present-day anthropogenic emissions there are large uncertainties that are not accounted for in this study. On the other hand, if the present-day anthropogenic simulations are used to investigate what the present-day aerosol would be like had there not been changes in BVOC emissions, a more

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relevant question would be what would the aerosol look like if there had been no land use changes (since temperature and CO2 changes did happen and are tightly tied to changes in anthropogenic emissions). However, there are no simulations available to answer this question.

I am therefore not convinced of the true value of the BE1.AE2 vs. BE2.AE2 simulations in the first place. At the very least, they should not be presented as the "baseline" against which other simulations are compared (abstract and section 4.2, perhaps also section 4.1 although there it is not stated what MEGAN runs are discussed!!!). A good candidate for the "baseline" would be BE1.AE0 vs. BE2.AE0, since this is also used in the radiative effect calculations and it would make MEGAN and LPJ-Guess runs directly comparable. This choice of a baseline could then be compared to BE1.AE0 vs. BE2.AE2 runs to estimate the relative impact of BVOC and anthropogenic changes. (The only potential issue would then be the XSOA runs, which are made with anthropogenic emissions, but I'm sure the authors can figure out a way to weave also these runs into the text so that they do not create confusion).

Minor comments:

1) p 26300, l. 2: "by absorption, scattering and reflection" – isn't reflection a subcategory of scattering (alongside with refraction and diffraction)?

2) p 26300, l. 15: "two dominant sources" – what other sources are there beside nucleation and primary emissions?

3) p 26300, l. 18: why ~80 nm? can vary greatly between different environments.

4) p. 26301, l. 29-> "because of the large uncertainties in these enhancements" – are the uncertainties any larger than in many of the other factors that you do take into account? It's fine for the scope of this study that anthropogenic effect on yields is not accounted for, but Iäm not convinced the uncertainties are the reason why they are left out.

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5) section 2: Why are decadal means used for MEGAN and annual means for LPJ-Guess? It is later stated that the BVOC emissions are sensitive to meteorological conditions; if this is true, using only annual avarages could severely bias the LPJ-Guess results at least in some of the regions and make comparison to MEGAN difficult.

6) p. 26302, end: I would argue that Acosta Navarro et al. 2014 shows very different (not somewhat different) magnitude of emissions from the two models

7) p. 26304, l 28: "predicted sesquiterpene emissions — predicted" – delete first 'predicted'

8) p. 26308: here AE2 simulations are outlined as the baseline runs; see major comment b) on why I don't think it is the best choice.

9) section 3.2: The large land-use changes discussed in the study mean that the surface albedo has not been constant between years 1000 and 2000. The same goes possibly also for cloud albedo due to regional climate changes. The authors should discuss the implication of these effects to their radiative effect calculations.

10) The description of the AIE calculation should be somewhat elaborated so that there is no need for the reader to refer to Scott et al. (2014). Where are the ES model unperturbed effective radii from? It seems that they are fixed (to what value?) – how realistic is this assumption? How realistic is the globally uniform updraft velocity? At what altitude are CDNC calculated (throughout the clouds indicated by ISCCP?)? How will these simplifications impact the calculated radiative effects?

11) Section 4.1: Indicate which MEGAN runs are discussed here. It would also be interesting to see some global mean values in addition to the regional values. Discussion of the LPJ-Guess results are quite vague; consider adding the same three panels for LPJ in Figure 2.

12) P. 26310, l. 22: what does "meaningful" mean in this context?; l. 25: "of this magnitude" – what magnitude?

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13) P. 26312, second half of the page: there is “firstly” and “thirdly”, but no second point.

14) p. 26313, l. 17: “such that there are more particles in the BE2.AE2.meg simulation” – more particles where?

15) p. 26313, bottom: “contrary to the previous case, with anthropogenic emissions turned off, —“ – the punctuation creates some confusion; does the “with anthropogenic emissions turned off refer to the “previous case” or to what follows? I assume the latter.

16) Section 4.2: I found this section heavy to read (large number of simulations, very long paragraphs). Consider restructuring the text into more digestible units by using shorter pragraphs, or even adding subsections for each of the sensitivity aspects.

17) p. 26318, l. 5: “This shows that anthropogenic land-use changes over the past millennium have decreased the number of CCN sized particles globally —“ – this is quite a strong statement (e.g. “indicates” would be better than “shows”) but possibly also inexact: 1) It is more likely that global CCN number has increased (due to anthropogenic activities); 2) The impact of land-use on CCN number is also uncertain, since land use changes have led to increased amine emissions (animal husbandry) which may have changed atmospheric nucleation as a CCN source in a complicated way.

18) Figure 10 and Table 2 give overlapping information. Consider whether the information could be presented in one or the other.

19) p. 25319, 1st paragraph (and throughout manuscript): you don’t simulate temperature change, so you cannot say “regions of cooling” or “band of warming” (“cooling/warming effect” is in my opinion ok). Whether a region would in reality experience warming or cooling depends also on several other climate forcers as well as changes in atmospheric and ocean circulation.

20) p. 26320, l. 18-20: Please elaborate how the combined aerosol radiative effect is calculated. It is not clear to me what “calculated simultaneously” means.

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21) p. 26322, l. 3-5: “on the order of 1 W/m2” – based on Figure 11, it is on the order of 0.5 W/m2, which is significantly less.

22) Section 4.3: You should also mention the radiative effect from anthropogenic emission changes (AE0 versus AE2) for comparison.

23) Section 4.4.: The general discussion on the model limitations is very good. However, I would like to see also some discussion on how the mentioned uncertainty sources are likely to affect the study’s conclusions.

24) Section 4.4., last paragraph is identical with the last paragraph of section 4.3.

25) p. 26325, l. 20: “with any certainty” is again quite a strong statement; consider reformulating.

26) Table 1: Explain abbreviations BE1, AE2, etc. also in the figure caption

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