Response to reviewer comments for the manuscript: **Aerosol size distribution and radiative forcing response to anthropogenically driven historical changes in biogenic secondary organic aerosol formation** by **D'Andrea et al.**

We would like to thank the anonymous reviewers for the overall positive and insightful comments on the manuscript. The original comments are in italics and the response to each comment is directly below the comment in bold. We will submit a revised version of the manuscript and figures with the changes outlined below.

**Response to review 1:**

*This study describes the response of SOA, and related radiative impacts, to millennial changes in BVOC emissions based on previous work by Acosta Navarro et al., 2014. The study is straight-forward and the paper is clearly presented. I have only minor technical comments and suggestions, detailed below.*

*1. Abstract, lines 1-12: These lines summarize results from a previous study and therefore do not belong in the abstract of this study.*

**We feel that some of this text is necessary to motivate our work; however, we have cut out roughly half of this text. It now reads, "Emissions of biogenic volatile organic compounds (BVOC) have changed in the past millennium due to changes in land use, temperature and $CO_2$ concentrations. Recent reconstructions of BVOC emissions predicted that global isoprene emissions have decreased , while monoterpene and sesquiterpene emissions have increased; however, all three show regional variability due to competition between the various influencing factors."**

*2. Pg 26299, lines 4-7: For completeness, the authors may wish to mention the important role that BVOC emissions (esp. isoprene) play as an O3 precursor.*

**We have included the following text into section 1 of the manuscript: "BVOCs are also important precursors for $O_3$ (Chameides et al., 1998) and secondary organic aerosol (SOA)..."**

*3. Page 26299, lines 21-22: the language "competing factors" and "anthropogenic factors" is a bit vague. It would be helpful if the authors could outline all the controlling factors, and perhaps define here which factors are considered as "anthropogenic" and which are included in this study. For example, the study considers the effect of CO2 fertilization and land use change, but not O3 damage to vegetation, another leading anthropogenic factor, and this isn't clear until the methods are presented.*

**We have included the following text into section 1 of the manuscript: "...due to competing factors such as land-use change, increases in $CO_2$ concentrations and temperature change. The most dominant cause of BVOC emission changes has been from anthropogenic factors (e.g. change in land cover and $CO_2$ effects)."**

*4. Page 26300, line 29: Jimenez et al., 2009 only report non-refractory measurements of aerosol (AMS); strictly speaking this is not "total mass".*

**We have changed "total mass" to "submicron particulate mass".**

*5. General: the paper makes inconsistent use of the oxford comma. See for example in the same paragraph line 17 vs. lines 25-26. Please harmonize.*

**We have gone through the manuscript and harmonized the oxford commas.**

*6. Page 26304, line 28: It seems that "approximately constant" would be a fairer characterization*
*than "increase overall" for a 1% change.*

**We have changed "increase overall" to "increase regionally, however remain approximately constant globally".**

*7. Page 26306, line 11: missing word "however, we will discuss. . ."*

**We have included "we" into the sentence.**

*8. Page 26307, line 9: errant reference typo? "(Lamsal et al., 2008)"*

**The reference has been included because Lamsal et al. (2008) show that ground-level $NO_2$ concentrations inferred from the satellite-borne Ozone Monitoring instrument are an order of magnitude less than concentrations found by Kroll et al. (2006).**

*9. Page 26307, lines 7-11: Note that while high absolute concentrations of any species may call into question the atmospheric relevance of chamber experiments, the NO:HO2 ratio within a chamber is an equally critical parameter for describing the chemical regime of SOA formation (i.e. fate of peroxy radicals).*

**We have included the following text into section 3.1 of the manuscript: "We note that while high absolute concentrations of any species may call into question the atmospheric relevance of chamber experiments, the NO:$HO_2$ ratio within a chamber is an equally critical parameter for describing the chemical regime of SOA formation."**

*10. Page 26307, lines 24-26: It would be useful if the authors could briefly summarize* *previous model evaluation of this particular simulation, since no comparison with observations is presented in this study.*

**We have included the following text into section 3.1 of the manuscript: "D'Andrea et al. (2013) evaluates GEOS-Chem-TOMAS particle number concentrations against measurements and shows that including the extra SOA yields improved number predictions for a wide range of particle sizes."**

*11. Page 26309, lines 17-22: What is the source of the properties used in these calculations (refractive indices, densities, hygroscopicities, etc)?*

**We have included the following text into section 3.2 of the manuscript: "The refractive index for each size section is calculated as the volume-weighted mean refractive index of the components (given at 500 nm in Table A1 of Bellouin et al., 2011), including water. Water uptake is tracked explicitly in GEOS-Chem-TOMAS by using ISSOROPIA (Nenes et al., 1998). For computational efficiency, the optical properties (dimensionless asymmetry parameter, and scattering and absorption coefficients, in $m^2\ kg^{-1}$) are then obtained from look-up tables of all realistic combinations of refractive index and Mie parameter (particle radius normalized to wavelength), as described by Bellouin et al. (2013). These aerosol optical properties were then included in monthly climatologies when running the offline ES radiative transfer model."**

*12. Page 26322, lines 10-11: this statement is missing a reference*

**We have included the following reference: Heald et al., 2009.**

References:

Bellouin, N., Rae, J., Jones, A., Johnson, C., Haywood, J. and Boucher, O.: Aerosol forcing in the Climate Model Intercomparison Project (CMIP5) simulations by HadGEM2-ES and the role of ammonium nitrate, J. Geophys. Res. Atmospheres, 116(D20), D20206, doi:10.1029/2011JD016074, 2011.

Bellouin, N., Quaas, J., Morcrette, J.-J. and Boucher, O.: Estimates of aerosol radiative forcing from the MACC re-analysis, Atmos Chem Phys, 13(4), 2045–2062, doi:10.5194/acp-13-2045-2013, 2013.

Chameides, W. L., Lindsay, R. W., Richardson, J., and Kiang, C. S: The Role of Biogenic Hydrocarbons in Urban Photochemical Smog: Atlanta as a Case Study, Science, 241, 1473–1475, 1998.

Heald, C. L., Wilkinson, M. J., Monson, R. K., Alo, C. A., Wang, G. and Guenther, A.: Response of isoprene emission to ambient $CO_2$ changes and implications for global budgets, Glob. Change Biol., 15(5), 1127–1140, doi:10.1111/j.1365-2486.2008.01802.x, 2009.

Nenes, A., Pandis, S. N. and Pilinis, C.: ISORROPIA: A New Thermodynamic Equilibrium Model for Multiphase Multicomponent Inorganic Aerosols, Aquat. Geochem., 4(1), 123–152, doi:10.1023/A:1009604003981, 1998.

**Response to review 2:**

*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 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.).*

**Radiative effects can be calculated between any two simulations to test what the radiative perturbation would be due some change in modelled emissions, process etc. For example, one may be interested in the radiative effect of switching all coal power plants to nuclear power plants: you'd want a simulation with present-day coal power plants to be your baseline (not pre-industrial). Pre-industrial simulations with little or no anthropogenic emissions are used as a baseline when looking at the radiative effects of the addition anthropogenic emissions (e.g. in the IPCC radiative forcings figure), and thus many simulations use pre-industrial as a baseline. However, pre-industrial by no means needs to be used as a baseline for radiative effect calculations.**

**In our work, we wanted to determine what the radiative effects were of changing biogenic emissions. In reality, the biogenic emissions changed at the same time anthropogenic emissions changed over the past 1000 years; however, we want to isolate the biogenic radiative effects from the anthropogenic effects (i.e. a partial derivative of radiative forcing to biogenic emissions changes). Because the presence of anthropogenic emissions would change the effect of changing biogenic emissions, throughout the paper we calculate the radiative effects of biogenic emissions changes with both anthropogenic emissions on and off.**

**That said, we were inconsistent in what we wrote in the abstract versus in section 3.4, so we have modified the abstract to be consistent with the discussion in 3.4. Thanks for pointing this out.**

*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.*

**Yes, they are unrealistic. But changing the biogenic emissions while leaving anthropogenic emissions off is equally unrealistic as neither actually happened. Both the biogenic emissions and anthropogenic emissions have evolved over the past millenium. We calculate the partial derivatives to biogenic emissions changes both with anthropogenic emissions on and off, and we necessarily needed to choose one to be the main focus in different sections. Fortunately, as is shown in Tables 2 and 3 and Figure 10, the response is qualitatively similar when anthropogenic emissions are on or off. We have added text in section 3.1 when we describe the simulations: "Thus, we estimate the effects of changing biogenic emissions in sets of simulations where the anthropogenic emissions are either on or off. While neither of these comparisons is realistic (anthropogenic emissions changed as the biogenic emissions were changing), it allows us to bound the impact of anthropogenic emissions on the partial derivative with respect to changing biogenic emissions."**

*b. Furthermore, I do not agree that comparing simulations with and without presentday 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.*

**What we meant to say is that the effects of changing biogenic emissions are different depending on whether anthropogenic emissions are on or not. We have reworded the text to say, "Additionally, the magnitude of the forcing of the biogenic changes differs whether anthropogenic emissions are on or off."**

*On the other hand, if the present-day anthropogenic simulations are used to investigate what the presentday aerosol would be like had there not been changes in BVOC emissions, a more 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).*

**While we disagree that the AE0 simulations are a better baseline (AE0 and AE2 are both equally limited), the simulations with anthropogenic emissions off are more direct comparisons with the LPJ-GUESS simulations. Therefore, the abstract has been changed to reflect this.**

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

**We have removed "reflection" from the sentence.**

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

**We have removed "dominant" from the sentence.**

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

**We have updated the manuscript to include a range of sizes (30-100 nm).**

*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.*

**It is still unclear what the magnitude of the uncertainties in the effect of changing anthropogenic pollution are on biogenic SOA yields. It is also unclear how to represent these anthropogenic uncertainties in the model, therefore these effects have not been explicitly investigated.**

*5) section 2: Why are decadal means used for MEGAN and annual means for LPJGuess? 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.*

**This is a good point. However, for LPJ-Guess we only received emissions for year-1000, year-2000, and the mean over the full time period. We have added the following sentence to Section 2, "Because our LPJ-GUESS emissions are from one single year at 1000 and 2000, these data may be susceptible to some regional biases due to not capturing interannual variability."**

*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*

**We have removed the following text from the manuscript "The two different models show similar trends but somewhat different magnitude of the emissions (see Figures 4 and 5 in Acosta Navarro et al. (2014))"**

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

**We have removed the first "predicted" from the sentence.**

*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.*

**See above comment.**

*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.*

**We have added the following text to section 3.2 of the manuscript: "Note that the land-use changes that lead to the changes in BVOC emissions explored in this paper may also lead to surface albedo and/or cloud changes. The would impact the Earth's radiative budget independently of the BVOC changes; however, we do not explore these changes in this paper."**

*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?*

**We have added/editted the following text in the third and fourth paragraphs of section 3.2 of the manuscript: "The cloud-albedo AIE is calculated by perturbing the effective radii of cloud droplets in the ES radiative transfer model. A control cloud droplet effective radius ($r_{e1}$) of 10 μm is assumed uniformly, to maintain consistency with the ISCCP derivation of liquid water path, and for each experiment a perturbed field of effective radii ($r_{e2}$) for low- and mid-level (below 600 hPa) water clouds are calculated as in Eq. (1) using the control ($CDNC_1$) and perturbed ($CDNC_2$) fields of cloud droplet number concentration for each month.**

$$r_{e2} = r_{e1} \times [CDNC_1 / CDNC_2]^{1/3} \qquad (1)$$

**We calculate monthly mean CDNC using the aerosol size distributions predicted by GEOS-Chem-TOMAS and a mechanistic parameterization of cloud drop formation from Nenes and Seinfeld (2003), for a globally uniform updraft velocity of 0.2 m $s^{-1}$. The assumption of a globally uniform updraft velocity is in itself a simplification and the AIE we calculate will be sensitive to the value used. Spracklen et al. (2011) and Pierce et al. (2013) found that assuming a base value of 0.2 m $s^{-1}$ gave an AIE close to the mean AIE obtained when the globally uniform updraft velocity was varied between 0.1 and 0.5 m $s^{-1}$. The cloud-albedo AIE is then calculated by comparing the perturbed (using $r_{e2}$) net radiative fluxes at the top of the atmosphere, to a control simulation (using $r_{e1}$)."**

*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.*

**We now specify that it is for the MEGAN simulations with baseline SOA yields. Section 4.1 of the manuscript has been edited to include the global mean values as well as the SOA formation from the LPJ-GUESS BVOC emissions. Also, Figure 2 has been**

**updated to include the same 3 panels of SOA formation for the LPJ-GUESS BVOC emissions.**

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

**We have included the following text in the manuscript: "Decreases/increases in SOA formation exceeding 50% would significantly…".**

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

**We have replaced "thirdly" with "secondly".**

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

**We have included the following text in the manuscript: "southern mid-latitudes in oceanic and deforested regions particularly."**

*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.*

**We have updated the manuscript for clarity: "However, contrary to the previous case, with anthropogenic emissions turned off globally averaged N40 also increased."**

*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.*

**We have updated the manuscript by breaking up some of the longer paragraphs into more manageable lengths.**

*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.*

**We have updated the manuscript to the following: "This indicates that anthropogenic land-use changes over the past millennium have decreased the number of CCN sized particles globally through changes in BVOC emissions, with regional changes..."**

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

**We acknowledge the overlapping of the information; however, we feel that presenting the information through tabulating numbers as well as visually in a figure are both necessary in order to thoroughly present the information to the reader.**

*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.*

**We have updated the manuscript to address this issue.**

*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.*

**We have added the following text to section 3.2 of the manuscript: "The DRE and cloud-albedo AIE are approximately additive, but to give a combined aerosol radiative effect, one must account for spatial overlap; therefore, a combined aerosol radiative effect is calculated by perturbing the cloud droplet effective radii and aerosol climatologies at the same time in the ES radiative transfer model, and comparing the net radiative fluxes to a control simulation in which neither is perturbed."**

*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.*

**We have updated the manuscript to 0.5 W m$^{-2}$.**

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

**We have added the following two sentences to Section 4.3: "While this global-mean DRE from biogenic emissions changes is smaller in magnitude than estimated anthropogenic direct radiative forcings (e.g. estimates of -0.85 to +0.15 W m$^{-2}$ in the most recent IPCC report (Boucher et al., 2013)), the DRE from biogenic emissions changes may be much larger, regionally." and "Similar to DRE above, the global-mean AIE from biogenic emissions changes is smaller than estimated aerosol indirect**

**forcings from anthropogenic aerosols (e.g. -0.3 to -1.8 W $m^{-2}$ in IPCC AR4 (Forster et al., 2007)), but again the regional AIE from biogenic emissions changes can be significantly larger than the mean.".**

*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.*

**We have updated the manuscript to include the following statement in section 4.4: "We expect the general spatial patterns to be robust, not necessarily the magnitudes."**

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

**We apologize for the duplication of text and have removed the paragraph from section 4.4.**

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

**We have updated the manuscript to the following: "...climate through SLCFs can be accurately determined."**

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

**We have included the following sentence to the figure caption for Table 1: "In the simulation naming scheme, "BE" refers to biogenic emissions, "1" refers to year 1000, "2" refers to year 2000, "O" refers to off, "meg" refers to MEGAN BVOC emissions, "LPJ" refers to LPJ-GUESS BVOC emissions, "up" refers to upper bound SOA yields, and "XSOA" refers to the inclusion of the additional 100 Tg (SOA) $yr^{-1}$."**

References:

Boucher, O., D. Randall, P. Artaxo, C. Bretherton, G. Feingold, P. Forster, V.-M. Kerminen, Y. Kondo, H. Liao, U. Lohmann, P. Rasch, S.K. Satheesh, S. Sherwood, B. Stevens, X. Y. Z.: Clouds and Aerosols, in Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by J. B. Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen and P. M. M. A. Nauels, Y. Xia, V. Bex, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA., 2013.

Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M. and Dorland, R. V: Changes in atmospheric constituents and in radiative forcing, in Climate change 2007: the physical science basis. contribution of working group I to the fourth assessment report of the intergovernmental panel on climate change, edited by S. Solomon, D. Qin, M. Manning, Z. Chen, M. Marquis, K. B. Averyt, M. Tignor, and H. L. Miller, pp. 129–234, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA., 2007.