Interactive comment on "Modeling anthropogenically-controled secondary organic aerosols in a megacity: a simplified framework for global and climate models" by A. Hodzic and J. L. Jimenez

Anonymous Referee #1

The intention of this paper is to provide a parameterization for SOA to be used in simplified models. The parameterization includes emission of an anthropogenic and biomass burning SOA parent hydrocarbon that is spatially distributed like CO and biogenic SOA from isoprene, terpenes, and sesquiterpenes. The role of acid-enhanced and anthropogenic-enhanced biogenic SOA are examined.

R1.1) Given the intention of the paper to provide a simplified framework and the ability of the parameterization to predict SOA it is recommended that the authors (a) refocus their paper on validation of the parameterization and (b) remove the conclusions regarding the lack of an anthropogenic control on biogenic SOA. The model setup seems too coarse/lumped to resolve any anthropogenic enhancement to biogenic SOA.

Response 1.1-a: we agree that the main focus of our paper is to develop and evaluate a new empirical parametrization for SOA modeling that can be applied for anthropogenically polluted regions. To further emphasize this focus, we have expanded the proposed parameterization to include not only the prediction of the SOA mass but also the evolution of oxygen-to-carbon (O/C) ratio. This quantity is of great interest for global modeling studies because it can be related to the hygroscopicity of organic aerosols (Jimenez et al., Science, 2009) and used to predict the organic aerosol influence on CCN activation in regional and climate models. The following text has been added to the manuscript to describe the O/C parametrization and the associated results:

"2.4 Empirical approach for predicting oxygen-to-carbon (O/C) ratios

In addition to parameterizing the SOA mass, we have also derived an empirical approach to compute the oxygen-to-carbon ratio for organic aerosols. The measured O/C ratio is a good indicator of the level of processing of the organic material. Being able to accurately predict O/C ratios in models is of great importance for determining the hygroscopicity of organic particles (Jimenez et al., 2009) and their effects on CCN activation and clouds. As previously, we have parameterized the ageing patterns observed in O/C ratio as illustrated in Figure 1b to fit the O/C observations reported for Mexico City and its outflow region (Dzepina et al., 2011). Typically, the O/C atomic ratios vary from very low (~ 0.1) for primary emitted organics, to levels close to 0.35-0.4 in freshly formed SOA, and up to 0.8-1 for aged air masses that contain more processed organic material. For this purpose the oxygen amount and its increase due to organic aerosol ageing were explicitly modeled in the OPT simulation, adding an additional transported variable into the model. The increase in O/C ratios in SOA particles due to ageing of the aerosol was parameterized according to the following equation: O/C = 1-0.6 exp(-A/1.5) where A represents the photochemical age of the airmass in days and 1.5 is the aging time scale

for Mexico City conditions. The O/C ratio for fossil POA is assumed to be equal to 0.1, whereas for the biomass burning value of 0.3 is used. "

line 340: "The evolution of O/C ratio was added to the OPT model and the simulation dedicated to evaluating the predicted O/C ratios against observations was performed from 16 to 25 March."

Two additional figures (Fig. 1b and Fig. 5) have been added with the following captions:
Figure 1: A schematic representation of the evolution of (a) the SOA/∆CO and (b) oxygen-to-carbon ratios as a function of photochemical age. For the SOA mass, the observed ratios (gray polygons) are taken from de Gouw and Jimenez (2009) and DeCarlo et al. (2010) for urban airmasses, whereas for the O/C ratios the observed ratios are those summarized in Dzepina et al. (2011). Curves represent the estimated ratios for OPT, SPR and ROB-like simulations for typical daily average OH concentrations found in Mexico City of $1.5x10^6$ molecules cm^{-3}.

Figure 5: Time series and associated diurnal profiles of O/C ratios as derived from the aerosol mass spectrometer measurement in downtown Mexico City (T0) and as predicted by the first order parameterization derived in this study for the OPT simulation from 16 to 25 March 2006. The results corresponding to the volatility basis set and already reported by Hodzic et al. (2010) are also plotted for reference (i.e. using the VBS parameterizations of Robinson et al., 2007 in blue and Grieshop et al., 2009 in green). The gray shaded areas and the red vertical bars indicate the variability associated with the observations and the OPT model respectively.

3.4 Evaluation of predicted O/C ratios

Figure 5 compares the simulated O/C ratios with estimated values derived from AMS measurements at T0. The observed O/C ratios range from 0.2 to 0.7 and display a strong diurnal variability with lower values in the morning when the primary emissions from traffic and other sources exert a major influence, and higher values during the daytime (~12-4pm LT) when more secondary oxidized matter is being formed by photochemistry. The simulated O/C ratios arising from the OPT simulation show a reasonable agreement with the observed levels both in terms of magnitude and temporal variability. In particular, the ageing of the organic material during the afternoon hours seems to be well captured by this simplified approach as illustrated by the average diurnal profile. The nighttime O/C values are often underpredicted in the model, which is consistent with the fact that CHIMERE tends both to overpredict POA (with low O/C=0.1) during nighttime due to inefficient nighttime boundary layer mixing and underpredict the contribution of biomass burning during the late afternoon or early morning smoldering fires. Both of these issues are discussed in detail in Hodzic et al. (2009).

The result of our simulation is also noticeably closer to the observations than the predictions for the original volatility basis set treatment (Robinson et al., 2007) that had substantially lower O/C levels as discussed in Hodzic et al. (2010) and as illustrated in Figure 5. Our results are however comparable to predictions of the Grieshop et al. (2009) volatility basis set approach shown in Hodzic et al. (2010) in which 40% oxygen gain was assumed for each oxidation generation of organic material. The processes leading to this rapid enrichment in oxygen within the first few hours of ageing are currently not understood from a mechanistic point of view.

Line 659: (v) We have also demonstrated that the evolution of oxygen-to-carbon ratios can be parameterized in a simple way, and used to predict the oxidation state of the organic material in this region. The O/C ratios and temporal evolution predicted using the simplified approach are in a slightly better agreement with observations than the results obtained by Hodzic et al. (2010a) for the full volatility basis set of Grieshop et al. (2009) that was assuming a 40% oxygen gain upon every oxidation of organic vapors.

Response 1.1-b: Also, it seems that our conclusions regarding the anthropogenic effect on biogenic SOA have been misunderstood. We did not assert that there is no such control in a general sense, and for the record one of us is a coauthor on two ACPD papers (Spracklen et al., 2011 and Heald et al., 2011) where a major missing global SOA source is attributed to anthropogenic enhancement of biogenic SOA. It is precisely for that reason that we were interested in evaluating that possibility for the Mexico City region. In the context of the current paper, our conclusion was very limited, stating that anthropogenic enhancement of biogenic SOA could not explain **the spatio-temporal patterns of observed OA in and around Mexico City during MILAGRO**, although it could contribute to the regional background. **We do not claim to conclude anything about the global importance of that source from the current paper, nor about its importance in other regions.** The explanation in this particular case is that biogenic precursor emissions are located in the coastal mountain chains, far from Mexico City (see Fig. 1b in Hodzic et al., 2009), and their mixing ratios in Mexico City are small. For this reason they cannot produce strong local enhancements of SOA in Mexico City starting in the early morning, which correlate with urban emissions and dominate the observed SOA concentrations.

Once this misunderstanding is cleared, our conclusion does stand: the spatio-temporal patterns that would be produced by an anthropogenically-enhanced BSOA source do not match the observations, and therefore such a source is not dominant **for this study only.**

To clarify these points and avoid misunderstandings by the readers, we have reworded the section 3.5.2 and the conclusion section to read:

section 3.5.2: "We recognize that the enhancement mechanisms applied here based on Spracklen et al., 2011 are a rather simplified representation of the anthropogenic influence in biogenic SOA formation. However, these same mechanisms result in large OA production when implemented in a global model (Spracklen et al., 2011), and for that

reason it was of interest to test their effects in the Mexico City region. Our results indicate that pollution-enhancement of biogenic SOA is unlikely to play a major role in the elevated OA in the Mexico City region due to the physical separation of the pollution and biogenic source regions. However biogenic SOA likely plays a role in the regional background OA, consistent with previous studies (Hodzic et al., 2009) and a quantitative analysis of fossil vs. non-fossil carbon in this region (Hodzic et al., 2010b). Future studies should address tthe question of anthropogenic enhancement of biogenic SOA using regional models and observations in regions where pollution and biogenic sources are in close proximity, and testing more detailed parameterizations e.g. by directly accounting for particle acidity effects."

conclusion: (vi) We have also examined whether the potential enhancement of biogenic SOA production under polluted conditions may explain the observed spatio-temporal patterns and levels of SOA in and around Mexico City. As parameterized here the SO_2-enhanced BSOA production is substantial. However this enhancement alone could not explain the spatio-temporal patterns of observed SOA in and around Mexico City, although it could contribute to the regional background concentrations. Our result does not imply that these interactions are not important in other regions or at the global scale, but that their relative impact is limited in this particular megacity that is dominated by anthropogenic and biomass burning sources.

Finally, in order to simulate the enhancement effect in biogenic SOA formation from anthropogenic sources we have implemented in our regional model study the same enhancement mechanisms proposed by Spracklen et al. (2011) at the global scale. We note that the rates and amounts of SOA formed by these mechanisms are not arbitrary, but were constrained to result in major global sources of the order of tens of Tg SOA/yr. We do agree with the reviewer that the parameterizations of Spracklen et al. (2011) are very simplified and that they have some shortcomings as pointed out by both reviewers in their further comments below. Because the main purpose of this study is not to study the anthropogenic-biogenic interaction, and because the influence of biogenic sources (located far from Mexico City) is limited in this region (as already demonstrated by our previous work in Hodzic et al. (2009) and in this paper), we have not carried out additional modeling work to improve the enhancement mechanisms proposed by Spracklen et al. 2011. However, we have discussed the limitations and possible improvements suggested by the reviewers in the revised manuscript, as summarized in response to comment R1.1. above.

R1.2) Comments regarding model set-up: The conclusion that an anthropogenic enhancement of biogenic SOA does not exist seems circular. The model input parameters are set in such a way that a biogenic enhancement may not be predicted. First, the emission of VOCA/CO is set to the observed ratio of SOA/deltaCO in aged air. This assumption implies that VOCA is the primary, if not only, source of SOA. The emission and aging of VOCA seem to be set in such a way that they should reproduce SOA concentrations, leaving little room for biogenic hydrocarbons to contribute. Secondly, the rate constants for reactions 11-14 in Table 1 may be set arbitrarily low. As these reactions are not fundamental kinetic reactions and are intended to

represent a coarse scale process, the rate constants are highly uncertain. If the rate constants for 11-14 are set low, then the model will naturally predict that there is no biogenic enhancement from the enhancement test. The importance of biogenic SOA may also be underestimated in the Lane et al. 2008 test since it reverts to older ISOP + OH low-NOx SOA yields.

Response 1.2: we disagree with the reviewer, as our more specific conclusion (as explained in response to R1.1) is **based on the spatio-temporal patterns** of the SOA concentrations in and around Mexico City, **and not on the absolute magnitudes**. Again, this does not mean that anthropogenic enhancement of biogenic SOA is not an important SOA source at other locations. As discussed in response to comment R1.1, we have modified the text to make these distinctions clear.

Regarding the constant rates, we again point out that our conclusions are based on the spatio-temporal patterns of the model vs observations, and not on the absolute amounts. However, the rates in our study are not arbitrary, we have simply adopted the parameterized mechanisms and rates that were evaluated by Spracklen et al. (2011) and used to derive conclusions on the importance of biogenic aerosols at the global scale. The rates and yields of Spracklen et al. were chosen to produce large sources of tens of Tg SOA/yr on a global scale, and thus are not "arbitrarily low".

Even if we had considered higher yields for BSOA production similar to the BSOA results of Hodzic et al., 2009 (based on Henze and Seinfeld, 2009 for isoprene and Pun et al., 2008 for other species), biogenic SOA would have been formed upwind of the city, and advected in the valley as aged background aerosols. This would have produced higher BSOA concentrations, but the diurnal profile would not have a strong photochemical signature as found in measurements, and as already discussed in Hodzic et al., 2009.

R1.3) Figure 5 along with other statements (pg 889) in the manuscript indicate that the acid catalyzed SOA pathway, as represented by homogeneous reaction with SO2, does not do a good job in predicting the spatial or temporal pattern of SOA. Although SO2 may be an attractive species for use in a global model since it is primary, the particle phase acidity should be better represented by the secondary product sulfate. Many global models have at least sulfate aerosol if not ammonium as well. This information would provide a better measure of particle acidity and may lead to better diurnal variation and spatial distribution.

Response 1.3: We agree that Spracklen et al. 2011 parameterization of this effect is very simplified, however as explained in response to comments R1.1 and R1.2 and in the text that has been added to the manuscript in response to those comments and quoted above. Mexico City does not appear to be a good environment to test more sophisticated parameterizations of these effects, due to the remote location of biogenic VOC emissions with respect to the pollution sources. Indeed, the biogenic SOA that is advected over the Mexico City valley as a background aerosol is already aged and processed, and the initial biogenic VOCs have mostly

reacted already. The reviewer's suggestion is useful, and we have included it in the paper as something to be explored in the future studies and other regions.

R1.4) (a) Missing information: The conclusion that the model does not show an anthropogenic enhancement of biogenic SOA should be removed.
(b) In addition to the comments about the reasoning being circular, a model that only matches observed concentrations within 30% may not have the skill to predict a biogenic enhancement of SOA. Furthermore, the model is missing known anthropogenic enhancements to biogenic SOA: 1. Table 2 shows the same SESQ SOA yields under high- and low-NOx conditions. Ng et al. 2007 have demonstrated that sesquiterpenes have higher yields under high-NOx conditions. 2. Table 2 shows that high-NOx isoprene photooxidation results in substantially less SOA than high-NOx conditions. New work indicates that isoprene SOA yields are the same, if not higher, under high-NOx conditions compared to low-NOx conditions [Chan et al. 2010].

Response 1.4-a: see responses to items R1.1, R1.2, and R1.3 above.

Response 1.4-b: in some of our simulations we have used SOA yields published by Lane et al., 2008, which have been applied in several modeling studies for this region e.g. Tsimpidi et al. (ACP 2010) and Shrivastava et al. (ACP 2011). We agree that these yields could be on low side and underpredict the amount of BSOA that is produced regionally. We used higher yields in our previous study (Hodzic et al., 2009), which did result in higher levels of BSOA background concentrations, mainly from isoprene.

However, and as discussed in the ACPD version, we also used the a BSOA treatment based on Henze and Seinfeld (2008) with yields ~5-10 times higher than for the parameterization of Lane et al. (2008). This is illustrated in Figure 5a (ACPD version) with the purple line. As explained above and as observed in the figure, using higher yields for biogenic VOCs will increase the background concentrations but won't reproduce the diurnal profiles of measured SOA, as biogenic emissions are distant with respect to the pollution sources, and BSOA is produced at the regional scale and advected into the city. As expected, the results using the parameterization of Henze and Seinfeld (2008) do not reproduce the enhancement in SOA levels that is occurring in the late morning.

In addition, the emissions of sesquiterpenes in the MEGAN model used here are two orders of magnitude lower than those of isoprene or one order of magnitude lower than those of alpha-pinene, and their contribution is expected to be small even as their yields are higher.

R1.5) Additional questions and comments. How do the authors reconcile measurements of modern carbon, which indicate Mexico City OA is 61-77% modern C, and model predictions that predict a small role for biomass burning and biogenic hydrocarbon SOA?

Response 1.5: we have in fact published a paper last year in this very topic:

Hodzic, A., Jimenez, J. L., Prévôt, A. S. H., Szidat, S., Fast, J. D., and Madronich, S.: Can 3-D models explain the observed fractions of fossil and non-fossil carbon in and near Mexico City?, Atmos. Chem. Phys., 10, 10997-11016, doi:10.5194/acp-10-10997-2010, 2010. http://www.atmos-chem-phys.net/10/10997/2010/acp-10-10997-2010.html

We can summarize the conclusions briefly here as: there are two datasets of non-fossil carbon for Mexico City during MILAGRO, which showed a systematic disagreement of ~15%, whose reasons remain unexplained. The fraction of non-fossil carbon downtown Mexico City of each dataset is 37-67% and 24-49%, respectively. Both datasets show an increase in the non-fossil carbon fraction of 15% when comparing high wildfire vs. rainy / low wildfire periods, which puts a bound on the contribution of that source, and is consistent with several other estimates (see also Aiken et al., ACPD 2010). We concluded that "Model results show that the relatively high fraction of non-fossil carbon found in Mexico City seems to arise from the combination in about equal proportions of regional biogenic SOA, biomass burning POA and SOA, as well as non-fossil urban POA and SOA" and that " he absolute modeled values of the non-fossil OC are consistent with the Swiss dataset but lower than the US dataset. Resolving the ^{14}C measurement discrepancies is necessary for further progress in model evaluation."

We have added a mention and citation of this study to the text, as summarized in response to comment R1.1. above.

R1.6) It is not clear in the title or abstract if the lumped SOA precursor is meant to include biogenic SOA. Conclusion #1 indicates that the proposed parameterization is an emission rate relative to CO and an oxidation rate of that precursor without any mention of how biogenic SOA is treated. Please state early in the manuscript whether the lumped precursor is intended to include biogenic SOA or not.

Response 1.6: the lumped SOA precusor is not meant to represent biogenic SOA. However, it may include some background biogenic SOA implicitly, since the origin of the parameterization is a fit to ambient observations. However, because the observations were predominantly in and around Mexico City, where the anthropogenic influence is largest, we think that any biogenic SOA included in the parameterization is a small fraction of the total SOA. We have clarified this in the revised paper with the following text in section 2.3:

"This parameterization has been derived using data from regions and plumes dominated by anthropogenic pollution. However, since it was derived by fitting ambient observations, it may include a small contribution from biogenic SOA to the regional background."

R1.7) Figure 4 is missing some a, b, c labels.

Response 1.7: fixed

R1.8) The purple line in Figure 5 is labeled as Hodzic OPT and Henze and Seinfeld 2006 yields. Please clarify.

Response 1.8: This is now explained in the caption of Figure 5:

“The purple line represents BSOA levels as calculated by the OPT run and using low-NOx yields for isoprene based on Henze and Seinfeld (2006).”

R1.9: Typo in title “controlled”

Response 1.9: fixed

Review by J. Pierce (jeffrey.robert.pierce@gmail.com)

This paper describes a simple approach of treating anthropogenic SOA and SOA associated with biomass burning in 3D models. For these SOA contributions, a parent SOA precursor is co-emitted with CO (with a constant emission ratio to CO) and oxidized to form SOA with a first-order reaction with OH. The authors also explore possible effects of anthropogenic species on biogenic SOA and conclude that while these effects can increase the biogenic SOA mass, they cannot reproduce the measured diurnal cycle of SOA. This paper provides a new alternative approach for modelling anthropogenic and biomass burning SOA, and the paper fits within the GMD goals. I believe that it should be published once the following comments have been addressed.

General comments

R2.1) I agree with referee #1 that there are weaknesses in the conclusions regarding the enhancement of anthropogenic species on biogenic SOA. Using the latest NOx dependent yields of biogenic SOA precursors that show larger yields in high-NOx conditions could be important. This should at least be discussed in the paper. I realize that the authors used SO2 as a proxy for aerosol acidity to be consistent with Spracklen et al. (2011). However, I would guess that SO2 and aerosol acidity are only weakly correlated with aerosol acidity near Mexico City. Since the model already predicts sulfate, ammonia and nitrate, it explicitly predicts inorganic aerosol acidity. While organics can modify this acidity, the inorganic acidity should be a much better predictor of aerosol acidity than SO2. I think it is unlikely that the effect of anthropogenic pollution on biogenic SOA could account for the diurnal cycle in organic mass; however, I feel that the above approaches would be an improved approach. These issues must at least be discussed in the revised paper.

Response 2.1: as discussed in response to comments R1.1-R1.3 from reviewer 1, we have clarified our conclusions about the anthropogenic enhancement of biogenic SOA, and the fact that our conclusions are specific (and robust) for this study, but that we do not make any broader conclusions. Regarding the acidity comment, we have added a suggestion for future studies but do not think that it is worth testing and implementing more complex models for our case study, as already described in response to R1.3.

R2.2) It was my impression that the IVOC distribution and the aging rates in Robinson et al. (2007) were first guesses with significant room for improvement. Thus, the Robinson approach could also easily be tuned to better match MILAGRO observations too. This should be discussed in the paper. Furthermore, the authors are already using a VBS approach for biogenic species, but is not discussed in their comparison of the number of species with the Robinson and Shrivastava formulations. Thus, I believe the computational benefits of the current approach are overstated relative to these previous works. I discuss this issue more in the specific comments.

Response 2.2: we agree that it would be possible for us to tune the Robinson et al. approach so that it matched the observations better. However a revised version of the Robinson

mechanism was published by the same authors in Grieshop et al. (2009), and in our previous work we already compared both parameterizations (Hodzic et al., ACP 2010; Dzepina et al., ACP 2009; Dzepina et al., ES&T 2011). A simplified version of the VBS parameterization has been recently published and applied to Mexico City and compared to some of the same data by Shrivastava et al. (2011). In addition a new two-dimensional version of the VBS is being developed to overcome some of the limitations of the 1D parameterizations and is undergoing extensive testing (Jimenez et al., Science, 2009; Donahue et al., ACP 2011, ACPD 2011; Murphy et al., ACP 2011). In the context of the different VBS parameterizations being proposed, we are not sure that a re-tuning of the published 1D-VBS to fit our observations and produce yet another parameterization is a useful project. Since the same functional dependence of the observations is already captured with our much simpler parameterization, we prefer to focus on that method, which can then be implemented in regional and global models.

Specific comments

R2.3) Page 872, lines 9-10: Multiple size bins is not necessary for aerosol mass predictions (even though it is used in this paper).

Response 2.3: We agree that the aerosol size distribution can be represented by either multiple size bins (sectional approach used here), or multiple modes (usually 3 modes). It is also possible to just calculate the bulk mass without size distributions, however that is rather uncommon nowadays for regional models and is becoming less common for global chemical transport models. We have modified our sentence to read:

page 3 - Introduction: "In addition, the full VBS approach is computationally very costly, as it requires keeping track of organic vapor and particle-phase species over 9 volatility bins and multiple particle size bins commonly used to represent the aerosol size distribution."

R2.4) Page 873, lines 1-20: How is the OA/delCO enhancement corrected for biogenic SOA?

Response 2.4: To derive this parameterization we consider the observed ratios in anthropogenically (or biomass burning) dominated regions i.e. megacity outflow areas, where the biogenic SOA production is small relative to the anthropogenic one. See Figure 3 of DeCarlo et al. (ACP 2010) which shows results from multiple studies, and also Figure 1 on our paper. Therefore, the contribution of biogenic SOA formation to the increase in observed OA/deltaCO ratios is assumed to be small.

This following sentence has been modified to specify that we are only dealing with the anthropogenic increase in SOA:

page 3 - Introduction: ***"The continuing good correlation between OA and CO and the large measured increase in OA/ΔCO ratios downwind of urban areas can then be attributed to the rapid growth of secondary organic aerosols due to anthropogenic pollution."***

R2.5) Page 880, line 3: Why is SO2 used as a surrogate for acidity? See general comment.

Response 2.5: See response to comment R1.3 from reviewer 1.

R2.6) Page 881, lines 9-19: In the discussion of the necessary number of species in the new model formulation, there is no mention of the biogenic species. You are already using a 4-bin VBS approach for this. Therefore, there is an additional 40 (9*4+4) species (58 total when including the 18 ASOA/BBSOA species), which gives you slightly MORE species than the Shrivastava approach where biogenic and anthropogenic organics are lumped together by volatility. Thus, I feel that the computational benefits of the current model configuration is overstated.

Response 2.6: We disagree with the reviewer on this point. Here we only compare the numbers for the treatment of anthropogenic and biomass burning species, and this new approach allows us to decrease the number of advected species from 162 to 4 species while maintaining or improving model performance, which is a substantial improvement.

Biogenic SOA is treated here separately and when the 4 bins VBS approach is used, we have an additional 36 biogenic species in the model (4*8 aerosol size bins). The biogenics are already lumped by volatility according to Lane et al., 2008, and it may be possible to lump them further into just two volatility bins as suggested by Shrivastava et al. (2010). However the simplification of the mechanisms of biogenic SOA formation should be the subject of further studies.

R2.7) Page 884, line 13: Please also cite Riipinen et al. (ACP 2011) and Pierce et al. (ACP 2011), which also show low volatility SOA using different methods than the other articles cited.

Response 2.7: we have added citations to both of those papers.

Pierce, J.R., I. Riipinen, M. Kulmala, M. Ehn, T. Petäjä, H. Junninen, D. R. Worsnop, and N. M. Donahue: Quantification of the volatility of secondary organic compounds in ultrafine particles during nucleation events, Atmos. Chem. Phys., 11, 9019-9036, 2011.

Riipinen, I., J. R. Pierce, T. Yli-Juuti, T. Nieminen, S. Häkkinen, M. Ehn, H. Junninen, K. Lehtipalo, T. Petäjä, J. Slowik, R. Chang, N. C. Shantz, J. Abbatt, W. R. Leaitch, V.-M. Kerminen, D. R. Worsnop, S. N. Pandis, N. M. Donahue, and M. Kulmala: Organic condensation: a vital link connecting aerosol formation to cloud condensation nuclei (CCN) concentrations, Atmos. Chem. Phys., 11, 3865-3878, 2011.

R2.8) Page 887, line 2: Does Spracklen et al. (2011) suggest that 90% of the predicted mass is biogenic origin near Mexico City or globally? Is it realistic to assume these two values would be similar? Please make this discussion more clear.

Response: the results of Spracklen et al. (2011) represent global averages, as discussed on that paper. However it is difficult to compare a global and a regional model, as suggested by the reviewer, and this is why we label this result as a "qualitative discrepancy" in the text that the reviewer is referring to. We feel that it is important to relate the conclusions of these two studies, although further follow-up studies will be necessary.

R2.9) Page 888, lines 4-5: "The difference is even larger for high NOx conditions and low organic mass (7 times)." What curve/panel are you referring to here?

Response: here and in the previous sentence we are not referring to any figure in the current paper, we are simply reporting ratios that we have calculated using both parameterizations. It seems to us that this text is already clear and we have not modified it.

Writing-related comments

R2.10) Title: "anthropogenically-controlled" doesn't need a hyphen because anthropogenically is an adverb, which already implies that it modifies controlled. It is not a joint adjective such as "human-generated SOA".

Response: fixed.

R2.11) Page 879, line 17, first sentence: It would be more clear to say "A similar approach as for anthropogenic SOA is adopted for biomass burning SOA formation". The previous paragraph discusses POA aging, not the ASOA formation via ratio w/ CO, so the way the first sentence was written was confusing.

Response: fixed.

R2.12) Page 884, line 22: "observation based"

Response: fixed

R2.13) Page 888, line 8: There should be a period after Pandis et al. (1991), not a comma.

Response: fixed

R2.14) Page 890, line 15: parameters should be parameter.

Response: fixed