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Atmospheric
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Discussions

Interactive comment on "The sensitivity of secondary organic aerosol (SOA) component partitioning to the predictions of component properties – Part 3: Investigation of condensed compounds generated by a near-explicit model of VOC oxidation" *by* M. H. Barley et al.

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Response to anonymous reviewer #2 on "The sensitivity of secondary organic aerosol component partitioning to the predictions of component properties – Part 3: investigation of condensed compounds generated by a near-explicit model of VOC oxidation" by Barley et al.

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The authors would like to thank the reviewer for their interest in the paper and their constructive comments to which we respond in detail below. New text to be inserted into the paper is shown in bold.

p. 21059 l. 2: *"A fairly constant diurnal profile in simulated ozone and degradation intermediates was simulated after 9 model days" How was the box model built to reach "the constant diurnal profile"? Is there a diurnal variation of the emissions, the photolysis, the temperature, the boundary layer height...? Is there any deposition or dilution of species included?*

The model was able to reach constant diurnal profile because there are temporal variations that have been applied to the NO_x and VOC emissions. The following text was added immediately before "A fairly constant diurnal profile..."

The diurnal profiles are based on the work of Jenkin et al., (2000) where single representative profiles describing seasonal, day-of-week and hour-of-day variations in emissions were assign to each of over 180 source categories in the NAEI. The model framework is that of a simple box model with no diurnal variation in temperature and boundary layer height. There is obviously no photolysis at night but there is deposition of major inorganic species and other species such as O_3. Long lived species were vented based on a boundary layer average lifetime of 3 days. This gave (the text continues) "a fairly constant diurnal profile in simulated ozone and degradation intermediates after 9 model days..."

p. 21059 l. 7: *"Further emission scenarios were simulated by independently multiplying the AVOCs, biogenic VOCs (BVOCs) and* NO_x *component of the base case emissions by factors of 0.01, 0.1, 10, 100 and 1000" Why is this range of variation selected? Is it representative of various conditions found in the UK or emission uncertainties?*

As is normally the case in sensitivity studies we chose a wide range in emission scenarios in order to explore as wide a chemical composition as possible. Obviously the extreme emission cases in the scenarios discussed in Section 3 do not represent typical UK atmospheric conditions (although they do encompass them) but that does not mean they should not/cannot be used to explore sensitivity of SOA formation to changes in emissions. The highest emission factors may be appropriate for down wind of a smoke stack while the lowest factors would occasionally reflect pristine west/south-west coastal environments.

p. 21059 l. 10: *"To study the trends ... relevant scenarios were used." If the simulations are not atmospherically relevant why are they used in Sect. 3 to discuss the sensitivity of SOA formation to emissions?*

We used the full range of emission scenarios to look at the trends in chemical composition and SOA properties described in Section 3. When moving to a more detailed analysis of the results it became appropriate to focus down, first on a set of 27 scenarios (eg. Figs. 2,7 and 8) that were more representative of typical UK emissions, and then, for analysis that required comparisons on a single case (eg. Figs. 3-6 and 9), a single scenario (1/1/1 NO_x-SF/AVOC-SF/BVOC-SF).

p. 21060 l. 4: *"The temperature and RH values were selected to cover the range of typical conditions found in a temperate maritime climate such as that of the UK" The relative humidity is set to 10, 30, 70 and 80*

We are not sure what the reviewer is asking here.

p. 21061: *How is the uptake of water taken into account in the model?*

The water partitions in the same way as any other component and contributes directly to the total condensed material (C_{OA}). The amount of water is determined by the selected relative humidity. In the non-ideal calculation the partitioning is solved iteratively with activity coefficients being calculated from the condensed phase composition using the group contribution method UNIFAC as described in the text.

To improve clarity the word "organic" will be deleted from p.21062 l.10.

p. 21064 l. 3: *The base case scenario should be introduced here. The level of* NO_x *and VOC simulated could be presented as well as the concentration of* O_3*, SOA mass, average O/C ratio, average N/C ratio, and average molar mass.*

The standard scenario (1/1/1 NO_x-SF/AVOC-SF/BVOC-SF) is first introduced in section

2.1 and the results from this scenario are first discussed in section 4.1 when Fig 3 is discussed. In Fig. 1 the standard scenario is at position (0,0) so we agree that we could provide more information about results using this scenario. Two sentences have been added to Section 3 providing the required information on the standard scenario:-

The standard scenario (1/1/1 NO_x-SF/AVOC-SF/BVOC-SF), first introduced in section 2.1 is found at (0,0) in Fig. 1. This scenario is run with 6219 ppt NO_x, 28828 ppt ozone and 31302 ppt VOC, of which 18231 ppt are due to methane. Predicted SOA mass (for the standard conditions) is 0.0547 $\mu gram.m^{-3}$; average O:C ratio is 0.9683, average N:C ratio 0.2112 and average molar mass 219.56.

p. 21064 l. 3: *"These plots are analogous to the conventional isopleths used to illustrate the dependence of ozone production on VOC and* NO_x*." The ozone isopleths usually represent the ozone concentration as a function of VOC and* NO_x *concentrations. Did the authors plotted in Fig. 1, Fig. S1 and Fig. S2 the logarithm of the concentration of VOC and* NO_x *(if yes, please give unity) or the logarithm of the factor applied to the emissions of the base case scenario? If this is the case, the authors should be clearer in the descriptions of the plots, the discussions and the legends and axis of the figures.*

We agree with the reviewer that this was not made clear in the paper. To avoid this confusion in terminology we have redefined AVOC and BVOC to be the primary emissions as measured in tons per year and introduce the AVOC or BVOC scaling factor (AVOC-SF or BVOC-SF) to describe the factor by which the average UK emissions (AVOC or BVOC) are multiplied by for a specific scenario. In Fig. 1, Fig. S1 and Fig. S2 it is the logarithm of the scaling factor applied to the emissions of the base case scenario that is used and the figures have now been amended to make this clearer.

p. 21064 l. 16: *"All three figures show similar trends with rising VOC and* NO_x*, suggesting a minimal dependence on the AVOC:BVOC ratio: i.e. a unit of AVOC is as likely to create SOA as a unit of BVOC" The comparison of the potential of AVOC and BVOC to form SOA could be very interesting...*

When we investigated this in more detail by comparing the quantitative amounts of SOA produced at the same scaling factors between the low AVOC-SF and low BVOC-SF case we found that the data supported "a minimal dependence on the ratio of the scaling factors" (ie. the ratio AVOC-SF:BVOC-SF), rather than the ratio of the input emissions.

...All three figures show indeed the similar general trends of SOA with changes in NO_x *or VOC. However, from the figures shown, I don't see how it can be concluded that there is "a minimal dependence on the AVOC:BVOC ratio" or that "a unit of AVOC is as likely to create SOA as a unit of BVOC" because : (i) if the yaxis are the logarithms of the factor applied to the emissions, the concentrations of VOC presented in the y axis are different between Fig. 1, Fig. S1 and Fig. S2. The concentration of VOC corresponding to log(10)VOC=3 in Fig. 1 is about equal to the one corresponding to log(10)Anthro=3 in Fig. S1 but about 10 times smaller than the one corresponding to log(10)Bio=3 in Fig. S2 (as in the simulated base case scenario, BVOC emissions are about an order of magnitude lower than AVOC emissions). (ii) the z axis color used for SOA mass in Fig. S1 and Fig. S2 doesn't look the same as in Fig. 1. It would help the discussion to have the same z axis.*

The reviewer is quite correct. The figures imply that there is a minimal dependence of SOA amount on the ratio of the scaling factors which suggests that biogenic emissions are some 8-9 times more effective in producing SOA than anthropogenic emissions. The phrase "a unit of AVOC is as likely to create SOA as a unit of BVOC" has been omitted and a new sentence has been added to Section 3:-

From p. 21064 l. 17:- "...suggesting a minimal dependence on the AVOC-SF:BVOC-SF ratio. **Due to the different levels of anthropogenic and biogenic inputs (see section), and assuming that the NAEI inventory accurately reflects the AVOC:BVOC**

ratio of emissions; this suggests that biogenic emissions are some 8-9 times more effective in producing SOA than anthropogenic emissions.
Finally the z axis scale has been made the same for the SOA mass plots in all three figures (S1, S2 and Fig 1).

p. 21064 l. 18: *The evolution of SOA mass as a function of VOC and* NO_x *level has been largely studied in chamber experiments (e.g. Kroll and Seinfeld, 2008). SOA isopleths as a function of* NO_x *and VOC have also been plotted in several papers (e.g. Vivanco et al., 2011, Camredon et al., 2007, Capouet et al., 2008). Most of the results show (i) an increase of SOA with increasing VOC concentration and (ii) two regimes of SOA formation depending on* NO_x *concentrations with an optimum reached at a given VOC/*NO_x *ratio. Here, the authors have to compare their results with previous literature. The authors should also try to use the model to explain the simulated* NO_x */ VOC limited regimes of SOA formation, together with the simulated average O:C, N:C and molar mass.*

All text from "a unit of AVOC is as likely to..." on line 17 to the end of this section has been deleted and replaced with an improved description of the NO_x dependence of SOA formation, including reference to the above works, and more detailed descriptions of the plots showing average O:C ratio, N:C ratio and molar mass. Reviewer 1 has also raised some concerns about Section 3. To answer the comments of both reviewers the entire section has been rewritten and two additional figures added to the Supplementary material. The rewritten Section 3 is reproduced in the response to Reviewer 1 with the new text highlighted in bold.

p. 21065 l. 1: *"Figure 2 shows the sensitivity of condensed SOA mass to the methods used to estimate vapour pressure and the treatment of solution ideality across all conditions for 27 model scenarios." Figure 2 also shows the sensitivity of condensed SOA mass to hydrolysis of acid anhydrides. Has hydrolysis also been applied for the scenario with low relative humidity?*

Yes. The lowest relative humidity we consider is 10%. It should be emphasised that the hydrolysed case was included to investigate the impact on predicted SOA of the inclusion of this single condensed phase reaction. While the equilibrium for the hydrolysis of anhydrides to the corresponding acids lies decisively on the side of the acid products the rates of the hydrolysis and the rates of condensation of anhydride from the gas phase would determine the degree of reaction. This may be much less than the 100% we have assumed and will be dependent upon the conditions. However at this stage we have no information on these rates so we have assumed 100% hydrolysis under all conditions.

p. 21065 l. 18: *Comparisons with previous published results on the influence of activity coefficient on SOA formation have to be performed (see for example Compernolle et al., 2009).*

The following text was added after "...Topping et al., 2011." on p. 21065 l. 18

The box-whisker plot for N-N/VP act in Fig. 2 shows that the inclusion of non-ideality can both increase and decrease SOA mass compared to the ideal base case. The results presented in Compernolle et al., (2009) show that the effect on yield of including non-ideality in the partitioning calculation can result in increased SOA mass under dry conditions but with the inclusion of water, and particularly at high %RH, the amount of SOA is reduced compared with the ideal calculations (see their Fig. 3). A more detailed analysis of our results to determine whether there was evidence of increased SOA mass at low RH and decreased SOA mass at high RH was not possible due to missing data for the non-ideal calculations resulting from the convergence issues described in Section 2.2.

p. 21065 l. 20: *Comparisons with previous published results on the influence of* *vapour pressure estimates on SOA formation have to be performed (see for example Valorso et al., 2011 and Compernolle et al., 2010).*

In Valorso et al.,(2011) the predicted SOA masses were substantially larger than the experimental values (in contrast to our results where the prediction for the standard scenario is some 20-100x too low) and although the JR-MY method predicts more SOA than the N-N/VP method the difference is about 12% for the low NO_x case, 50% for intermediate NO_x and a factor of x4 for the high NO_x case (see their Figure 5); rather that the couple of orders of magnitude seen in Fig. 2. The two sets of results are not directly comparable as those from Valorso et al., use a fully explicit, automated chemical degradation mechanism (GECKO-A) and the experimental systems being modelled are chamber studies with relative high concentrations of VOC (giving 5-30 $\mu gram.m^{-3}$ of SOA). There is some evidence from our results that for the higher emission scenarios (of the 27 used to generate Fig. 2) the factor difference between vapour pressure by JR-N/VP and by N-N/VP drops to less than x10 suggesting that the sensitivity to different vapour pressure methods decreases with increasing SOA mass.

p. 21065 l. 24: The following sentence has been added after "...two Joback methods (JR-N/VP and JR-MY) show a significant bias towards increased mass." **Similar results have been reported by Valorso et al., (2011) where the JR-MY method consistently predicted more SOA than the N-N/VP method although the differences were much smaller than those shown in Fig. 2**.

p. 21065 l. 28: The following sentence has been added after "...underestimating the slope of the vapour pressure curve (Barley and McFiggans 2010)". **Compernolle et al., (2010) also noted the large differences in estimated vapour pressures when using the MY and the N/VP vapour pressure equations, finding that the N-MY method under predicted their experimental SOA amounts while the N-N/VP method gave much better results.**

p. 21066: *The order of the ranking of the top 200 species has been plotted in Fig. 3 and S3 to look at SOA sensitivity to estimated vapour pressures, activity coefficients and hydrolysis. It would be useful to report the contribution of these 200 species to the total SOA mass. Are a few species among the top 200 species needed to represent the total SOA mass or are all these species dominant but present in SOA at a low concentration?*

p. 21066 l. 23: The following sentences have been added after "...confirming that the results seen in Fig. 3 are typical." **The contribution of the top 200 compounds to the SOA is $>$99 mole% for all the models considered. The distribution of the SOA species is very uneven so the top 2 compounds typically contribute 20-45% and the top 10 compounds some 50-75%. The model including the hydrolysis of acid anhydrides is exceptional in that the predicted SOA is dominated by two compounds (maleic acid and methylmaleic acid) which contribute 77.8 mole% to the SOA; the next 8 components contribute 10.9%. Hence the predicted SOA composition is dominated by a small number of compounds with a long tail of compounds that make a vanishingly small contribution to the aerosol.**

p. 21066 l. 21: *"It is not surprising that hydrolysis of anhydrides does not greatly change the compound ordering as the vast majority of compounds have the same vapour pressure as they have in the base case calculation." Indeed the hydrolysis of anhydrides does not substantially change the compound ordering for the conditions simulated in Fig. 3. However large differences are observed in the vapour pressure distribution shown for the same simulation in Fig. 4, with most of the SOA components in the logC*=-2 bin for the simulation including hydrolysis...*

In Fig 4 the bar graphs for the base case and the hydrolysed case are identical for logC*$<$ -2 (note change of scale). The big increase in the bar at logC* = -2 for the hydrolysed case is due to the formation of two new compounds (maleic acid and methylmaleic acid) from the corresponding anhydrides that are usually found in higher bins (logC* = +2 or more).

...Why is this difference not observed in Fig. 3? Is it because the hydrated compounds are not present in the top 200 species of the base case simulation?...

Yes- the corresponding anhydrides are too volatile to occur in the top 200 compounds of base-case SOA. Maleic anhydride occurs at position 211 and methylmaleic anhydride at 216. A sentence has been added to the paper explaining this:- continuing on from p .21066 l. 25 "...majority of compounds have the same vapour pressure as they have in the base case calculation. **The top two compounds in the hydrolysed SOA do not appear in Fig. 3 because the corresponding anhydrides are too volatile to be in the top 200 compounds for the base case SOA.**

It would be useful to clarify this in the manuscript and to specify, among the 200 species plotted in Fig. 3, the number of species that are identical between the base case simulation and the simulations performed to test the sensitivity of SOA.

p. 21066 l. 20-21: The following text has been added after "...involving hydrolysis of acid anhydrides, cause a substantial reordering of the compounds." **For a compound to appear in Fig. 3 it must be in the top 200 compounds for both the base case and the model used to test the sensitivity of SOA. The number of compounds that satisfy this requirement provide a metric for the degree of reordering of the compounds (high value minimal reordering; low value more reordering). Hence for Fig. 3 the degree of reordering increases in the order:- Hyd 189, SB-N/VP 180, N-MY 168, SB-MY 164, and JR-MY 160, JR-N/VP 147 and N-N/VP act 126.**

p. 21068 l. 1: *"If the hydrolysed case is compared to the base case then it is clear that the hydrolysis process reduces the concentration in bin logC*i=+2 and possibly some further bins of even higher volatility, while significantly increasing the amount of material in bin logC*i=-2." It can also be noted that the bin logC*i<-5 is largely reduced when hydrolysis is implemented...*

This is incorrect. The logC* $<$-5 bin is about the same size in the hydrolysed case and the base case (4.1 vs. 4.03 x10^7 molecules. cm^{-3} respectively). It is the change of scale in the y-axis that makes the bar look so small in the hydrolysed case.

*Does this mean that most of these species in this bin are formed from the oxidation of species that can be hydrolysed? If these species are hydrolysed, they are mainly in the aerosol phase and cannot be oxidized in the gas phase to give species in the logC*i<-5 bin. The contribution of these hydrolysed species to the total SOA mass could therefore be important. What is the contribution of the hydrolysed species to the total SOA mass?*

The species in the logC*$<$-5 bin are the same in both the base case and the hydrolysed case and are present in almost the same amounts. There is a slight increase in the amount of some species condensing in the hydrolysed case because of the increased aerosol mass (up from 0.0547 to 0.2045 μgram.m^{-3}). Other than this small increase in condensation the compounds in logC*$<$-5 bin are not affected by the hydrolysis process. The contribution of the hydrolysed species to the total SOA is indeed very important (increasing the condensed mass by a factor of 4) but this is almost completely due to two compounds in log C*=+2 bin moving to the logC*=-2 bin due to the reduced vapour pressure upon hydrolysis. For the case used in Fig. 4 these two compounds (maleic acid and methylmaleic acid after hydrolysis) provide 77.8 mole% of the SOA.

To provide greater clarity about the y-scales in Fig. 4 the following note has been added to the caption:- **please note scale change half way up the y-axis in each panel.**

p. 21069 l. 2: *"If the right hand side of Fig. 6 (molar mass above 200 amu) is compared to the corresponding figure (Fig. 9c) in McFiggans et al. (2010) substantial similarities can be seen confirming that differences in predicted SOA composition with* *vapour pressure estimation techniques are not completely systematic and independent of functionality." I don't understand what the authors are trying to say.*

We agree that this needs to be made clearer. Figure 5 shows the accumulated abundance of the components contributing to SOA binned according to their molar mass and their O:C ratio. The left hand figure shows this for the case where vapour pressures are calculated using the base case (N-N/VP) and the right hand figure shows a similar plot with vapour pressures by JR-N/VP which gives significantly more mass. Figure 6 shows the ratio of the accumulated abundances in the same array of bins. If the extra condensing mass had the same properties (in this case molar mass and O:C ratio) as the material predicted to condense using the base case then Fig. 6 would be a uniform colour (constant factor across all bins). The fact that it is not uniform shows that changing the vapour pressure method creates a bias in the other properties of the SOA:- in this case the molar mass and O:C ratio averaged over all the SOA move to lower values.

The text from p. 21068 l. 28 "The similarity..." through to p. 21069 l. 6 "...and independent of functionality" will be deleted and replaced with:- **If the extra condensing mass had the same properties (in this case molar mass and O:C ratio) as the material predicted to condense using the base case then Fig. 6 would be a uniform colour (constant factor across all bins). The fact that it is not uniform shows that changing the vapour pressure method creates a bias in the other properties of the SOA:- in this case the molar mass and O:C ratio averaged over all the SOA move to lower values.**

p. 21069 l. 23: *The author should discuss the results of Fig. 7 before comparing with McFiggans et al., 2010.*

The comparison with Fig. 10 in McFiggans et al., has been moved to the end of the section describing Fig. 7.

p. 21069 l. 24: *"As expected this plot has a similar form to Fig. 10 in McFiggans et al. (2010)," What do the authors mean by "a similar form" and why were the authors expecting to get a similar form? For a same case, nor the simulated median values of the molar mass or the O:C ratio, nor the spread of the box plots, are the same in the two figures. Only the relative evolution of the median values between all the sensitivity cases is comparable. What do we learn from this comparison?*

The comparison of Fig. 7 in the present paper with Fig. 10 in McFiggans et al., (2010) has been rewritten to better describe the similarities and differences between the figures. As both figures are derived from the condensation of large numbers of multifunctional molecules of diverse chemical structure over a wide range of conditions we would expect similarities in terms of the relative position of the simulated median values and similar relative variation in the size of the box-whisker plots. It would be expected (for example) that both plots would show median O:C ratio and median molar mass for the JR-N/VP method to be lower than the corresponding values for N-N/VP method. If we can demonstrate similarities between the two figures this would suggest that such features may be general for any large set of multifunctional molecules of diverse chemical structure rather than being a specific feature of a given set of molecules.

Starting at p. 21069 l.16 the section describing Fig. 7 has been rewritten with a substantial reordering of the old text and the addition of some new text to address the criticisms above. In the first sentence the words "...is directly comparable to Fig. 10 in McFiggans et al., (2010) showing... " have been deleted so that the sentence now reads:- "Figure 7 shows the difference and variability in the average O:C ratio and molar masses of the predicted condensed **SOA with different models across 27 scenarios (all combinations of scaling factors 0.1, 1.0 and 10)**". It then continues... "The black asterisk (**and** associated **black** box-whisker plot) shows..." incorporating the text from lines 18-23 without change. Lines 24-27 (that is from "As expected this plot has a similar form..." through to "...in Fig. 10 of McFiggans et al., 2010") are deleted and the last paragraph (p. 21070 l. 9-16) of this section is inserted at this point with a minor modification:- "In Fig. 7" is dropped so the text continues "The differences **in the median values** between the base case and the hydrolysed case...".

This is then followed by new text:- "**This plot can be compared to Fig. 10 in McFiggans et al., 2010. In Fig. 7 the median values of both O:C ratio and molar mass are clearly lower for the methods using the JR T$_b$ estimation methods. In Fig. 10 of the earlier paper the corresponding difference is less clear although the JR-N/VP does come out with the lowest O:C ratio.**" The text then continues from p. 21069 l. 27 "**In Fig. 7** the median values are at higher O:C ratio (0.76 compared to..." and continues to the end of the section (p.21070 l.16).

p. 21071 - 21072: *Comparisons with previous published results on the influence of activity coefficient on SOA formation have to be performed (see for example Compernolle et al., 2009).*

The results presented in Compernolle et al., (2009) show that the effect on yield of including non-ideality in the partitioning calculation can result in increased SOA mass under dry conditions but with the inclusion of water, and particularly at high %RH, the amount of SOA is reduced compared with the ideal calculations. This is consistent with many compounds showing negative deviations from ideality at low %RH but then moving towards positive deviations from ideality in the diluter solutions formed at high % RH.

The following text has been added to the paper at p. 21072 l.6 after "at all relative humidities. **This is consistent with results reported by Compernolle et al., (2009) which show evidence for both salting in and salting out of SOA components with increased salting out at high %RH.**

3. Technical corrections

p. 21059 l. 9:*"to give 206 emission scenarios" Should it be 216?*

10 scenarios were found to give atmospherically unreasonable ozone levels and were initially screened out giving a total of 206 rather than 216 scenarios. The missing scenarios also impacted upon the form of the figures(S1 and S2) in the Supplementary material due to missing data. On further investigation we found that these 10 scenarios did not adversely change the nature of the plots shown in the Supplementary material (surfaces extend monotonically into the affected region- no discontinuities) and have now been included in the full set. The "206 emission scenarios" highlighted by the reviewer has been changed to "216 emission scenarios".

p. 21064 l. 3:*"The properties are logarithm of the average condensed mass (μg m^{-3}); average O:C ratio; molar mass and average N:C ratio." Should it be "condensed mass" instead of "average condensed mass" and "average molar mass" instead of "molar mass"? See also the legends and axis of the figures.*

The reviewer is correct and these changes will be implemented throughout the paper.

p. 21064 l. 12:*"Figure S1 in the Supplement shows variability in the properties for the 35 scenarios with the lowest biogenic input (0.01); S2 shows them for the 35 scenarios with the lowest anthropogenic input (also 0.01)." Should it be 36 including the base case?*

Figures S1 and S2 have been updated using a full set of 36 scenarios as described in our response above (p. 21059 l. 9). The reference to the "35 scenarios" used in figs. S1 and S2 has been changed to 36 scenarios.

p. 21064 l. 15: *"All averages were calculated for the same atmospherically relevant case within a scenario T =293.15 K,..." It should also be specified that the SOA results are shown for vapour pressures calculated with N-N/VP and for the ideal case. See also the legends of the figures.*

Additional text has been added to make it clear that the calculations were done using the base case vapour pressure with liquid phase ideality; this has also been added to the figure captions.

p. 21065 l. 5 *"across all conditions for 27 model scenarios" It would clarify the section to say that the 27 model scenarios were with emissions multiplied by factors of 0.1, 1.0 and 10.*

The following text has been inserted after "across all conditions for 27 model scenarios":- **(all combinations of scaling factors 0.1, 1.0 and 10)**

Figures S1 and S2: Why are there no simulated data at high VOC and high NO_x*?*

As described above (response to comment re- p. 21059 l. 9) one datapoint was missing from the high VOC, high NO_x region in each of Figures S1 and S2 due to atmospherically unrealistic high ozone levels. This has now been corrected and the complete figures are in the new version of the Supplementary material.

Too many references are made to McFiggans et al., 2010. It should be possible to read the two papers independently and most of the time, this reference is not useful and confusing for the reader. For example, the following references can be removed: p. 21062 l. 16 : "As described by McFiggans et al. (2010)" p. 21065 l. 5 "This figure is directly comparable to Figs. 3 and 4 in McFiggans et al. (2010)." p. 21069 l. 16 "directly comparable to Fig. 10 in McFiggans et al. (2010)"

We agree that the last two are redundant and should be removed. The reference to the part 1 paper in p. 21062 l. 16 is in the methodology section and forms a link with the previous paper for the definition of the base case. We would like to retain this reference.

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