

Responses to reviewer 1.

Review of “A modeling approach to evaluate the uncertainty in estimating the evaporation behaviour and volatility of organic aerosols” by Fuentes and McFiggans.

We thank the reviewer about the useful insights and suggestions to improve this manuscript. Below we provide answers to general and specific comments (blue-Reviewer, black-Authors)

Summary: Fuentes and McFiggans report on the development of a model of particle evaporation in a thermodenuder. They use this model to analyze various measurements from the literature and perform various test computations in an attempt to provide insights into the operation and use of thermodenuders for the determination of aerosol volatility. What is new here is the attempt to relate their kinetic model to the empirical model of Faulhaber et al. (2009), and this is a useful contribution, although at times the discussion could be clearer and more precise. However, overall I unfortunately find that this manuscript covers little new ground, as much of the general concepts have already been discussed in the literature. For example, it has already been established that interpretations of thermodenuder measurements are sensitive to model inputs (including ΔH_{vap} and the evaporation coefficient). It has also previously been shown that re-condensation is particularly sensitive to mass loading coupled to time allowed for re-condensation. I strongly recommend that the authors refocus the entire manuscript to primarily discuss the new findings related to the interpretation of the empirical Faulhaber et al. (2009) method.

The reviewer's main concern is that he/she considers that the manuscript should be refocused on new findings on the analysis of volatility, and less attention should be paid to the re-condensation analysis. Although we agree that the most interesting area of our manuscript is the interpretation of calibration curves and volatility, re-condensation is considered in our study as a potential source for uncertainty which affects subsequent estimations on volatility; therefore the analysis on re-condensation should be shown in a first instance for presenting the results in a consistent manner. Furthermore, we disagree with the reviewer that the re-condensation analysis that we are presenting is not novel enough. The reviewer points out that previous literature already shows the effect of different parameters on thermograms. This is true for the evaporation process but not for re-condensation, which is the focus of the thermogram analysis in our study. To the authors' knowledge the only parameter whose effect has already been analysed regarding re-condensation is the aerosol mass loading (Cappa (2010)). However, the influence of parameters such as volatility, evaporation coefficient, diffusion coefficient and particle size on re-condensation, has not been shown before. Also, Cappa (2010) evaluated re-condensation by comparing thermograms at the end of the cooling section, with and without a charcoal denuder; however, the change of thermograms with respect to the outlet of the heating section, which is what induces deviations when inferring volatility from thermograms, has not been previously shown. With this parametric analysis we also provide additional detailed information on the conditions that may promote re-condensation. For example, we add new findings that re-condensation could be negligible at high aerosol mass loadings for samples with low accommodation coefficient. We agree that the abstract and conclusions of the manuscript should be modified to enhance that the main motivation of our work is to analyse the uncertainty in deriving volatility from thermograms by using different methods; but we also think that the detailed analysis on re-condensation is worthy to be presented for the reasons stated above. Thus, rather than re-structuring the whole paper, we consider that it is more appropriate to reduce and tighten the re-condensation analysis, so that only the main results are shown, and re-focus abstract and conclusions on our analysis on volatility.

Specific comments:

1. P. 6724, L. 12: Unclear what is meant by “thermograms may be lowered...”

This part of the sentence will be replaced by “thermograms may be altered”.

2. P. 6724, L. 25: This paragraph is difficult to follow.

The paragraph will be rephrased.

3. P. 6726, L. 8: Saleh et al. have shown that equilibration time is insensitive to compound vapor pressure, counter to what is stated here.

We disagree on this point. Although the characteristic equilibration time is not explicitly dependent on the saturation concentration, as shown in the dimensionless analysis by Saleh et al. (2011), the equilibration time is ultimately dependent on the saturation concentration through the dependence of particle size on volatility. The equilibration time is independent of the volatility “on the assumption that the change in particle size upon evaporation is small” (page 578, Saleh et al. 2011). A change in the particle size is, however, inherent to the evaporation process itself; hence, deviation from the above assumption is expected in most practical cases. This can clearly be seen in Figure 6 of Saleh et al. (2011), which shows that the equilibration time increases with increasing volatility. Hence, we do not consider that it is rigorous to state that the equilibration time is not dependent on the compounds volatility.

4. P. 6728, L. 7: Saleh et al. (2011) simply state that recondensation is negligible when the dimensionless coupling number C_n , which describes the ratio between diffusive mass transfer to particles to the convective mass transport to the walls of the tube, is <0.7 . It is stated in that work that under “most” conditions, this should be satisfied. Their C_n is related both to mass loading and the specific denuder design (in particular the diameter of the cooling section). There is no conflict between the conclusions in Cappa (2010) and Saleh et al. (2011) if one recognizes that the systems considered in these two studies had different geometries. The point of Saleh et al. (2011) is that one can design a TD where re-condensation will be minimized, but that one can also design a TD where re-condensation will be problematic. The claims in this manuscript that these two studies stand in contrast to each other is incorrect.

We agree with the reviewer that C_n values in the TD system employed by Cappa (2010) (eg. $C_n=3.5$ in Figure 5 by Cappa (2010)) were above the maximum value established by Saleh et al. (2011) for negligible re-condensation and, therefore, there would not be disagreement between these studies in this regard. However, there is still discrepancy between Cappa (2010) and Saleh et al. (2011) regarding their considerations on the potential for re-condensation. While Cappa (2010) stated that the re-condensation potential in the cooling section depends on the local conditions at the walls (i.e. local equilibrium versus non-absorbing boundary conditions), Saleh et al. (2011) claimed that re-condensation is negligible for most thermodenuder designs and experiments. This is because Cappa (2010) considered upper and lower estimates for re-condensation derived from these two boundary conditions, while Saleh et al. (2011) adopted the assumption of equilibrium wall condensation, i.e. the lower estimate. This will be clarified in the revised manuscript.

5. P. 6730, L. 25: Detailed balance requires that γ_{evap} and γ_{cond} are equal, if equilibrium is ever to be possible. As such, an assumption of different γ 's for evaporation and condensation is implicitly assuming that the system can never reach equilibrium; this should be made clear.

We disagree that different kinetic coefficients for evaporation and condensation imply that equilibrium can never be achieved. An assumption of different coefficients for evaporation and

condensation implies that the kinetic rate at which these two processes will tend to equilibrium will be different. If sufficient time is allowed for evaporation or condensation to occur, equilibrium will be attained regardless of the value of kinetic coefficient.

6. Equation 3: the "(CS)" subscript is confusing in terms of meaning.

This will be clarified.

7. Equation 4: It should be noted that the boundary condition implied by this equation is somewhat contentious. For example, Khlystov and co-workers have argued that it is inappropriate and that, in fact, the walls will be in equilibrium with the sample (at least if experiments are run for sufficiently long times, which may not be appropriate for ambient measurements but can be for laboratory experiments). See the discussion by Khlystov as part of Cappa (2010): <http://www.atmos-meas-tech-discuss.net/2/C1209/2010/amtd-2-C1209-2010.pdf> or the comment posted by Khlystov on this manuscript.

Our previous calculations using a boundary condition of no mass transfer to the walls provide for an upper estimate of the potential re-condensation in the cooling section. We agree with comments by Dr. Khlystov that an equilibrium wall condensation boundary condition, which represents a lower estimate for re-condensation, should also be considered, as the condition of no mass transfer to the walls may not always be realistic. As suggested, we have made recalculations including this condition with $C_{g,i}(\text{wall})=x_i C_{\text{sat},i}(T_{\text{wall}})$ (see attached figure at the end of this response (Figure **R1** and **R2**)). In Figure **R1** we present our previous results for upper estimate and new results for lower estimate of re-condensation. Our new calculations show that although the particle growth is lessened as a result of gas condensation on the walls of the cooling section, there is still significant re-condensation occurring at high mass aerosol loadings, with respect to the exit of the heating section. This affects our definition of the minimum aerosol mass loading for which re-condensation may be significant (i.e. $RF > 20\%$), with a new value of $50 \mu\text{g}/\text{m}^3$ versus our previous value of $30 \mu\text{g}/\text{m}^3$; however, this does not change our conclusion that re-condensation is substantial at high aerosol loadings for the TD design under study, even for the lower estimate implied by the assumption of wall condensation equilibrium conditions.

The inclusion of this condition also affects our results on the performance of the charcoal denuder to avoid re-condensation. Figure **R1** in this response shows that, under the assumption of equilibrium wall condensation, a charcoal denuder is of no use in reducing re-condensation for compounds of low volatility. As in Saleh et al. (2011), this is due to the gas concentration C_g being much higher than $C_{\text{sat}}(T_{\text{wall}})$, thus leading to similar gradients between the gas phase and the walls in the cooling and denuder sections (i.e. $C_g - C_{\text{sat}}(T_{\text{wall}}) \sim C_g - 0$). We have found that using a charcoal denuder is of no benefit for volatilities up to $1 \mu\text{g}/\text{m}^3$. For volatilities above this value (Figure **R2**), the charcoal denuder becomes slightly more efficient for avoiding re-condensation, however it induces particle evaporation below 45°C . As mentioned above, these results represent a lower limit for re-condensation, as the equilibrium conditions at the walls may not be achieved in all cases; thus, the charcoal denuder may still be useful at conditions where equilibrium on the walls is not attained.

The assumption of wall condensation also lessens the significance of re-condensation in affecting the volatility distributions derived from thermograms for a-pinene SOA in section 7. We have redone the cooling section calculations including this boundary condition in sections 4 and 5 of our study and reanalysed the volatility distributions which were affected by re-condensation in section 7. As Dr. Cappa, we consider that a condition of equilibrium at the wall is an assumption that will not necessarily be attained, and that both lower and upper estimates should be considered to derive the potential for re-condensation. We will present the results for upper estimate (no absorption to the

walls) and lower estimate (wall condensation equilibrium) of re-condensation in the revised version of the manuscript. We will include a new Figure in the supplementary material showing lower estimates for re-condensation at different aerosol mass loadings (Figure R1, bottom) and will also add curves for lower estimates of re-condensation in the parametric analysis of the revised manuscript and the final plots showing volatility distributions. Because some figures already include a large number of curves, we will not add extra data on lower estimates to all graphs (eg. Fig. 8 and 9); however, we will revise the text where the lower estimate may affect our interpretation of thermograms. In those cases in which only the upper estimate is presented, this will be clearly stated in text and figures.

The significant re-condensation degree that we have obtained for the lower estimate of re-condensation is a result of the geometry and experimental conditions of the thermodenuder (TD) designs simulated in our study. The dimensionless number C_n in our parametric simulations ranged from 2.7-10, which is well above the maximum value of $C_n=0.7$ established by Saleh et al. (2011) for negligible re-condensation. Hence, there is no disagreement between our results and those in Saleh et al. (2011). In our study we have worked with geometries and conditions representative of some currently used TD designs, such as Huffman et al. (2009), which presents C_n values much higher than 0.7 at high aerosol loadings (eg. $C_n=2-7.4$ for 100 nm particles at 400 $\mu\text{g}/\text{m}^3$ and $C^*=0.01 \mu\text{g}/\text{m}^3$). Although we acknowledge that TD geometries could potentially be modified to reduce the effect of re-condensation, the aim of our study is to analyse the problems derived when using existing TD systems, even if their geometries are not optimum to minimise re-condensation issues.

8. P. 6732, L. 25: I do not follow this argument. In particular, what does “constant number profile” mean? Constant number concentration? Or constant absolute number?

This paragraph will be revised. By “constant number profile” we mean “constant particle concentration profile”.

9. P. 6736, L. 2: The Epstein et al. equation is not “linear”.

The term “linear” will be removed.

10. P. 6736, L. 8 and Figure 3: If the authors are really attempting to span a range from “ambient” to “laboratory”, the lower limit considered here should be much lower than 50 $\mu\text{g}/\text{m}^3$. A value more like 1-5 $\mu\text{g}/\text{m}^3$ seems more appropriate to me.

We tried to span a range from ambient to laboratory conditions that was significant for the re-condensation analysis. Because re-condensation is not significant for aerosol loadings below 30-50 $\mu\text{g}/\text{m}^3$, we do not see necessary to study the behaviour for aerosol loadings below these values.

11. P. 6736, L. 23: Although this conclusion may be true (that re-condensation can still be important even at “low” mass loadings), it should perhaps be noted that it is unlikely that anyone would actually include such a long residence time “cooling” section if there was not a charcoal denuder. So, in practice this may not be appropriate. In other words, people would likely rapidly switch to ¼” tubing (or the metric equivalent) instead of wider diameter tubing following the heating section.

Although it is reasonable to shorten the length or reduce the diameter of the cooling section in order to reduce the residence time and minimise re-condensation, for practical reasons it is not always possible to do this to a minimum that ensures that re-condensation will be negligible. For instance, in the design of Huffman et al. (2008), a cooling section length of 15 cm (15 s. residence time) was necessary for the sample to cool down to ambient temperature (Figure 2 of our manuscript). Furthermore, reducing the cooling section diameter would require extending the

length of the tubing to allow the sample to cool down, so this measure might not be as helpful as thought. Also, sometimes due to configuration limitations particular to field measurements or when injecting the thermodenuder exhaust into chambers, long tubing lengths may be required. It should also be noted that some researchers work with commercial thermodenuders that already incorporate a cooling/denuder section. It is less likely that these groups will modify the original configuration of their equipment and they should be made aware of the potential issues implied. A brief discussion on the practical aspects of our conclusions will be included in the revised version of the manuscript.

12. P. 6737, L. 1: This conclusion seems a bit premature, before the influence of the denuder on evaporation of particles that have higher volatility is explicitly considered. This is only the case here because the authors have used a relatively “low” volatility compound for their test ($C^* = 0.01\mu\text{g}/\text{m}^3$).

This conclusion will be modified to account for the results of our recalculations with a boundary condition of wall condensation. Also, as suggested by the reviewer, this conclusion will be placed later on, after the volatility analysis is presented.

13. P. 6737, L. 18: The authors should clarify that they mean ambient multi-component mixtures that are (likely) composed of significant fractions of compounds with $C^* < 1\mu\text{g}/\text{m}^3$. However, it is possible to easily create laboratory mixtures where compounds with $C^* > 1\mu\text{g}/\text{m}^3$ are highly abundant. And, in fact, if equilibrium partitioning theory applies, the fraction of material that is “high” volatility should increase as the total mass loading increases, making the potential for induced evaporation to increase as mass loading increases in multi-component mixtures.

Agreed. This will be clarified with the following statements: “Because the significance of this effect is limited to a narrow range of temperatures and volatility compounds, it is likely that the impact of denudation on thermograms is of low significance for ambient multicomponent mixtures. For laboratory experiments at high aerosol loadings, the mass of high volatility compounds will be significant and the evaporation effect induced by the denuder may be more important than for ambient measurements.”

14. P. 6737, L. 26: I do not see what makes this a “remarkable” result. This is exactly as expected.

Here we meant “noticeable”. This word will be replaced.

15. P. 6738, L. 7: I believe it is the change in particle surface area, not number, that is most important here.

The reviewer is correct that it is the total particle surface that directly influences re-condensation. We will modify this sentence.

16. Figure S3: It is difficult to see the base case separate from the two other cases considered. The Figure will be modified for clarification.

17. Figure 6: It is difficult to compare subtle differences between figures (that may end up on different pages in final MS). Please include at least one curve from Figure 5 on Figure 6 for reference. Further, the figure caption is insufficient, as it is not stated what the dashed vs. solid lines or the different colors correspond to.

Reference curves will be added to Figure 6 and legend will be completed.

18. P. 6739, Line 7: The authors never actually report what vapor pressures and enthalpies of sublimation they have assumed for succinic acid. Values in the literature vary, and without specifying what values were used the derived accommodation coefficients are without meaning because they cannot be placed in the appropriate context. Values in the literature range over more than an order of magnitude for the VP of this compound. In other words, without better constraints on the actual vapor pressure (and ΔH_{sub}) for this compound, the accommodation coefficient cannot be determined with any precision whatsoever from this analysis.

The value of C_{sat} and ΔH_{sub} used for the calculations were the averaged values from data provided by Chattopadhyay and Ziemann, (2005), Bilde et al. (2003) and Davies and Thomas (1960). We will add information on these data in the revised manuscript. Dr. Khlystov argued in his interactive comment that our analysis could be pre-determined by the vapour pressure values used for the calculation, in case that the method used for deriving vapour pressures was based on an assumption of unity for the accommodation coefficient. Chattopadhyay and Ziemann (2005) and Bilde et al. (2003) employed a TPTD and a TDMA method, respectively, in which the accommodation coefficient was assumed to be 1. This assumption has been shown to be of low significance in TDMA studies, with variations of the accommodation coefficient in a range between 0.2 and 1 leading to changes in the vapor pressure estimation in less than 30% (Bilde et al. (2003)). In the work of Davies and Thomas (1960) the vapor pressure was determined by means of an effusion method, without any assumption on the value of the evaporation. The value derived from Davies and Thomas (1960) is in fact very similar to that from Bilde et al. (2003) (P25 of 4.23×10^{-5} and 4.6×10^{-5} Pa, respectively). Thus, it is not evident that the accommodation coefficient value derived in our work was pre-determined by the vapour pressure values used as input to the model. We believe, however, that further work is necessary in order to clarify differences between the accommodation coefficient values from different studies. We will add this discussion to the revised manuscript.

19. P. 6739, L. 21: The way this is phrased the authors seem to be implying that the measurements by Cappa and Wilson (2011) were for dicarboxylic acids, which is not the case.

This sentence will be clarified.

20. P. 6740, L14: I think this conclusion is somewhat misguided in that it does not account for how TD's are actually designed. If a charcoal section is included, the TD will likely be designed to have a "long" residence time, whereas if no charcoal denuder were included, people would be foolish to allow a long residence time. Instead, they will likely minimize the time between the heating section and the measurement in their design. I believe that this sort of reality check on the likely design of a given TD should be discussed explicitly.

As explained in response **11**) although it is reasonable to shorten the length or reduce the diameter of the cooling section in order to reduce the residence time and minimise re-condensation, for practical reasons it is not always possible to do this to a minimum that ensures that re-condensation will be negligible. For instance, in the design of Hufmann et al. (2008), a cooling section length of 15 cm (15 s. residence time) was necessary for the sample to cool down to ambient temperature (Figure 2 of our manuscript). We agree that an explicit discussion on the practical aspects of our findings should be included in the manuscript.

21. Figure 8a: Why are the computational results at the lowest temperatures not shown? Instead, the calculations seem to begin at the second lowest temperature.

Calculations were run for temperatures starting at 35°C for no particular reason.

22. P6741,L13: It is entirely unclear what is meant "...fed with the same lubricating oil aerosol sample...". What is being fed? What values were used? It is also not clear how this analysis or conclusions differ from that of Cappa and Wilson (2011)?

We will rephrase this whole paragraph. Here we mean that the same lubricating oil aerosol sample was used in the TD and chamber experiments of Grieshop et al. (2009), however we show that the accommodation coefficient derived from their TD experiments was 0.3 (in agreement with TD measurement by Cappa and Wilson (2011)), while their chamber measurements yielded an accommodation coefficient of 0.001-0.0001. The slow evaporation rate of particles in Grieshop et al. (2009) dilution chamber may have been induced by the release of material from the chamber walls.

23. Page 6741, L 10: A general comment on citations in AMT: I personally believe it is not appropriate to have "Interactive Comments" listed as "references". I find this to be somewhat misleading as it suggests that such comments have been peer reviewed. (Similarly, I have a fundamental problem with people citing articles that have been "published" in APCD and AMTD. These are not published papers. They are submitted manuscripts. There is a difference, as too often seems to be forgotten. Obviously, this is a comment not specific to this manuscript.) I suggest that it should instead be made clear in the text that this point is being made based on a non-peer reviewed source. Perhaps "(Grieshop, Interactive Comment on Cappa and Wilson, [http:..., 2011](http://...))" is more appropriate.

We will do as suggested.

24. P. 6742, L. 20: An interesting finding, if true. However, the authors miss an opportunity to suggest a simple way to determine whether this is the case or not: perform experiments at lower mass loadings or with bigger particles where re-condensation becomes less important. Additionally, it is interesting to note that this type of analysis (different uptake and evaporation coefficients) provides an upper limit on the evaporation coefficient when the uptake coefficient is assumed to be unity. (What is meant specifically by "close to unity"?)

We believe that this is an interesting suggestion for future experimentation that should be included in the revised version of the manuscript. We will clarify that assuming a condensation coefficient of 1 implies an upper limit for the evaporation coefficient. By "close to unity" we mean ≈ 1 .

25. P. 6746, Line 14: "estates" should be "states". This will be corrected.

26. Figure 16: The way the Pathak curves are presented makes it appear as if there is mass below $\log C^* = 10^{-2}$. This is not the case, as these should be zero. I suggest that the figure should be changed to make these appear as zero (i.e. have no red showing at all).

Unluckily the graphing software that we use does not allow for the bars of zero value not to be showing as a flat line.

27. Figure 11: Although I understand why there should be a dependence of the $1/T_{50}$ values on mass loading, I do not understand why the y-axis values ($\log C^*$) are changing in the different panels. This, to me, seems wrong since C^* is a fundamental property of the compound being simulated (i.e. is a model input). Perhaps I am misunderstanding, but from the discussion and figure caption it is not clear.

These plots seem confusing because we have not represented the same volatility compounds for the different aerosol loading cases. For instance, alkane nC30 is represented in the plot at $10 \mu\text{g}/\text{m}^3$, but

it is not present in the plots at 150 ug/m³ and 400 ug/m³. We reckon that this leads to confusion since it looks as though C* was modified when in reality it is 1/T₅₀ that is shifted between the aerosol mass loading cases. To make this clear we will represent the same volatility points in each of these plots.

28. Discussion regarding C* and T₅₀: The Faulhaber et al. (2009) relationship provided a quick and dirty way to estimate C* from measurement of T₅₀. The results presented here suggest that the exact relationship is subject to many caveats, which is very useful to know. However, I am not convinced that the results are presented in the most useful manner. It is stated clearly that the results here relate only to “equilibrium thermograms.” This is fine, as far as it goes, but it is also known that it takes very long for systems at low mass loadings to actually achieve equilibrium. Thus, when the authors show, for example, results at C_{OA} = 10 μg m⁻³, they are assuming an equilibrium that likely will never be obtained in an experiment. I think that it is a useful exercise to consider the limitations of the Faulhaber method, but I am not convinced that Figure 11 is useful. However, Figure 12, which accounts for non-equilibrium in the TD, seems more appropriate/useful to me. (Note: the α = 1 case in Figure 12 is not easy to distinguish or identify from the legend.) I suggest that the authors revise this entire section.

We believe that Figure 11 should be kept in the manuscript so that the limitations of the Faulhaber et al. (2009) empirical equation and its theoretical meaning can be understood. We will clarify in the manuscript that the case of C_{OA}=10 μg m⁻³ in Figure 11 represents equilibrium that will not be achieved in real conditions in a thermodenuder. We will revise Figure 12 and text in this section.

29. Unlike Khlystov et al. in their comment on this manuscript, I do not agree that MFR is always “problematic for characterizing aerosol volatility.” Yes, MFR depends on aerosol mass loading, but as discussed in this manuscript and elsewhere, appropriately used kinetic models can aid in the interpretation of MFR curves. The suggested use by Khlystov et al. of the “change in mass concentration across the thermodenuder” is certainly a useful alternative metric, but this requires knowledge of “ΔC_{max}”, which is the maximum change in aerosol concentration and corresponds to equilibrium conditions. However, as pointed out in this manuscript and in Saleh et al. (2011), under some conditions certain systems are unlikely to reach equilibrium and thus determination of this ΔC_{max} may prove problematic as well in practice.

We agree with the reviewer’s comment.

30. Discussion of dilution on p. 6750: *I don’t find any of this to be surprising. If the authors assume the aerosol evaporates slowly (small γ values), of course it takes a long time to reach equilibrium. Further, shouldn’t the results oscillate with time for conditions when the evaporation and condensation γ values are assumed to not be equal? Finally, why is a condition where γ_{evap} = γ_{cond} = 1 not considered? Is this essentially equivalent to the equilibrium case?*

These results may not be surprising but they give a useful indication on the magnitude of aerosol mass overestimation/underestimation under the assumptions of kinetic-controlled conditions or equilibrium defined by the partitioning theory. We do not understand why the reviewer believes that there should be oscillation when the coefficients are set not to be equal. On plot 13 (a) we represent the formation of aerosol as a result of cooling by condensation and plot 13 (b) represents the evaporation process due to dilution. We state both the evaporation and condensation coefficients in the legend to make clear whether we assume that condensation is affected or not by the amorphous solid state of α-pinene SOA; however, we use only one of the coefficients for the calculations (i.e. condensation coefficient for plot 13 (a) and evaporation coefficient for plot 13(b)). The case with glassy state affecting both evaporation and condensation is that with equal

coefficients ($1e-4$), while the case with condensation not being affected by the particle phase is that with condensation coefficient=1. The condition at equilibrium is that for both coefficients=1 for the time scales considered or any coefficients as long as enough time is provided to achieve equilibrium. We will clarify these aspects in the text.

32. P. 6753, L4: If the SOA really is “glassy” and the evaporation coefficient is very small, does it make sense to think that condensation will proceed with unit accommodation coefficient? Wouldn’t the “glassy” nature of the SOA inhibit (re)condensation as well?

The slow evaporation of α -pinene SOA aerosol has been attributed to the barrier to the diffusion process due to the amorphous structure of the particles. Because gas re-condensation is a surface process this may not be affected by the particle internal phase. In such a case, the kinetic coefficient for re-condensation could exhibit a higher value than that for the evaporation process. It should be noted that, because the kinetic limitation in reality may lie in the diffusion in the condensed phase through a viscous particle, the evaporation coefficient does not need to equal the condensation coefficient. This was already explained in the paper when deriving the accommodation coefficient values from α -pinene SOA thermograms (P. 6742, L. 3-15).

33. P6754, L25: I do not understand the reference to Topping and McFiggans (2011). To me, this particular reference adds little to the discussion and seems to be a random example.

In this paragraph we briefly mention the type of studies that could be affected by the uncertainty associated to the volatility distributions derived from thermograms. Not only studies on the global distribution of organic aerosols are based on these volatility distributions, but also new analysis on the role of the organics volatility properties on aerosol cloud activation through co-condensation. We think it is interesting for the reader to know about these types of studies and a reference is included for illustration.

34. P6754: Determination of volatility distributions from evaporation measurements (i.e. thermogravimetric measurements) are of course dependent on the assumed mass transfer coefficients. However, this does not necessarily translate directly into uncertainties in models so-long as self-consistent treatments are used. In other words, if one assumes $\gamma = 1$, then there is a specific volatility distribution that corresponds and should only be used if one continues to assume $\gamma = 1$. If a different γ is assumed, then a different volatility distribution is obtained. These two parameters are linked, and thus models that recognize this and make sure to use these parameters self-consistently may not exhibit as many problems as implied by this discussion.

We agree that models can be self-consistent if a given accommodation coefficient and its corresponding volatility distribution are used systematically. However, the quantitative results of models will primarily depend on the assumption of accommodation coefficient, thus, even if a model is used in a self-consistent manner, its outcome will be subject to the uncertainty associated to the value of this coefficient and its corresponding volatility distribution. We will clarify this in the discussion.

35. P6754, L5: It should be noted that the inability to fit the α -pinene SOA mass thermogram with the model when $\gamma > 0.01$ is only true when the specific ΔH_{vap} distribution here is used. If the H_{vap} distribution is changed (for example, using smaller values of ΔH_{vap}), it may be possible to obtain model/measurement agreement for larger γ values.

We agree with this statement. In our model we have assumed that H_{vap} is a function of C^* , following Epstein equation. If H_{vap} presents different values to those defined by Epstein equation a

different range of alpha values would be possible. We will add this statement to the revised manuscript.

36. Throughout the authors use the term “glassy” aerosol. It is not clear that such particles are actually glassy. A more appropriate and general term would be “amorphous solid” or “amorphous semi-solid”, after Koop et al. (PCCP, 2011, 13, 19238).

The term “glassy” will be replaced by “amorphous solid”.

References

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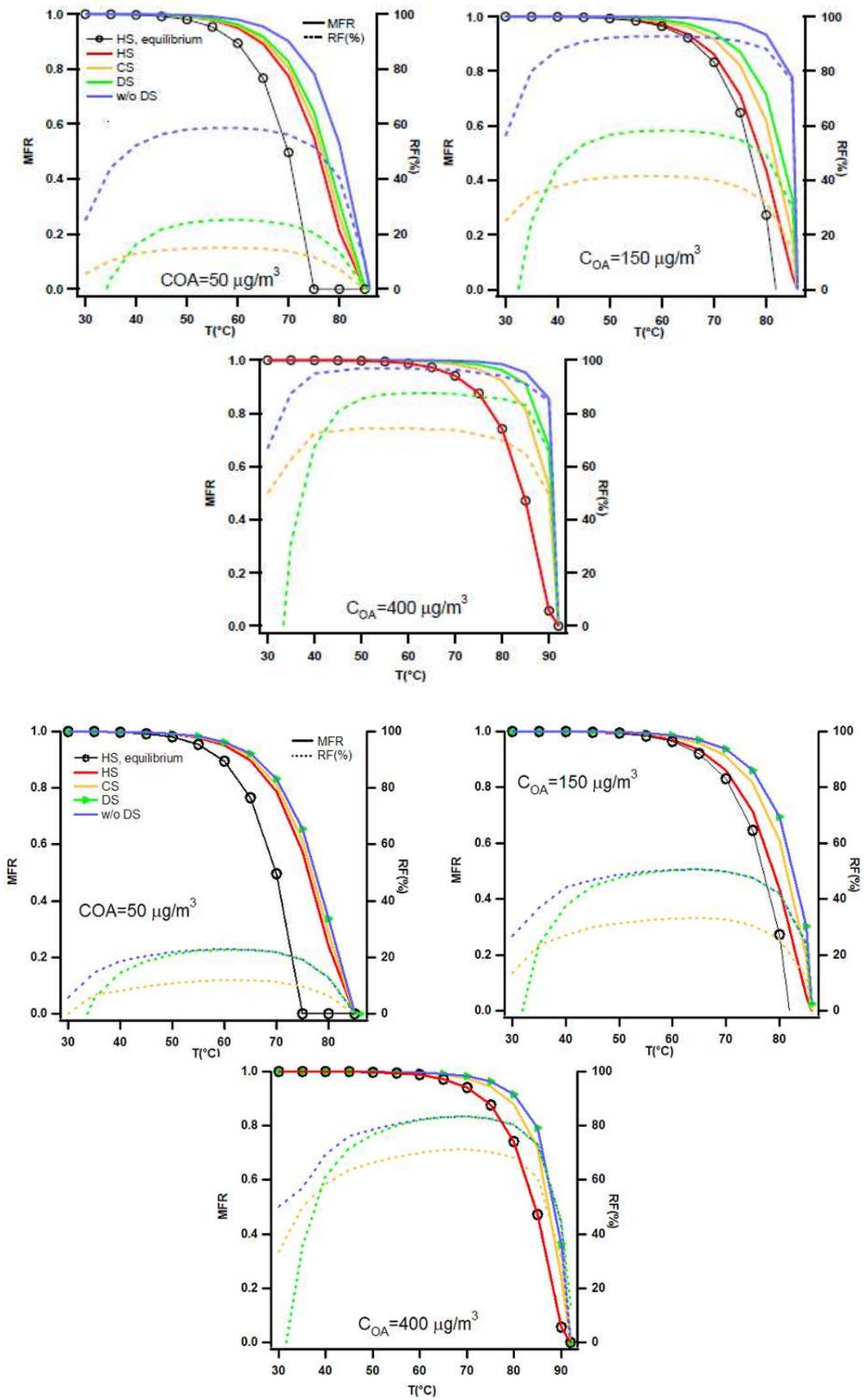


Figure R1: Upper (top 3 figures) and lower (bottom 3 figures) estimates of re-condensation as a function of aerosol mass loading. Plots represent output thermograms (MFR) and re-condensation fraction (RF) for the heating section (HS), cooling section (CS), denuder section (DS) and equivalent configuration without denuder section (w/o DS). Baseline case: $C_{sat} = 0.01 \mu\text{g}/\text{m}^3$, $d_p = 100 \text{ nm}$, $D_i = 5 \times 10^{-6} \text{ m}^2/\text{s}$ and $a = 1$.

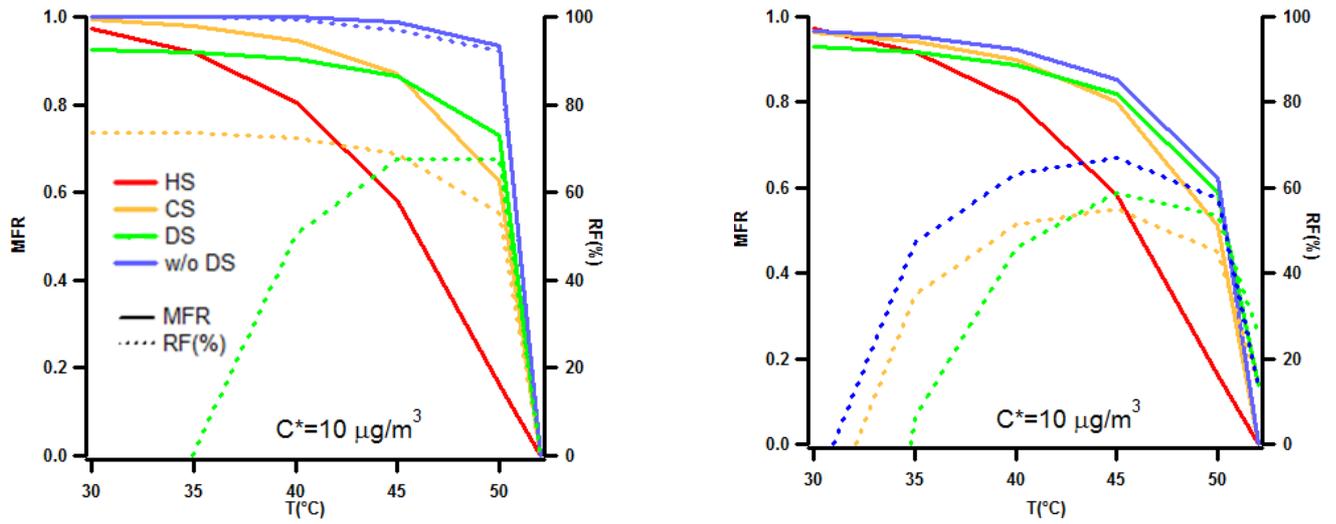


Figure R2: Upper (left) and lower (right) estimates of re-condensation for the case with $C_{\text{sat}}=10 \mu\text{g}/\text{m}^3$, $C_{\text{OA}}=400 \mu\text{g}/\text{m}^3$, $d_p = 100 \text{ nm}$, $D_i=5 \times 10^{-6} \text{ m}^2/\text{s}$ and $a=1$. Plots represent thermograms and re-condensation fraction for the heating section (HS), cooling section (CS), denuder section (DS) and equivalent configuration without denuder section (w/o DS).

