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

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 γ, 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.

Specific Comments:

1. P. 6724, L. 12: Unclear what is meant by "thermograms may be lowered…"
2. P. 6724, L. 25: This paragraph is difficult to follow.
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.
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.
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.
6. Equation 3: the "(CS)" subscript is confusing in terms of meaning.
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.

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?

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

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 $\mu g/m^3$. A value more like 1-5 $\mu g/m^3$ seems more appropriate to me.

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.

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 $\mu g/m^3$).

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 $\mu g/m^3$. However, it is possible to easily create laboratory mixtures where compounds with C* > 1 $\mu g/m^3$ 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.

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

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

16. Figure S3: It is difficult to see the base case separate from the two other cases considered.

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.

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.

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.

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.

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.

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)?

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)" is more appropriate.

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"?)

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

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).

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.

28. Discussion regarding C^* and T_{50}: The Faulhaber et al. (2009) relationship provided a quick and dirty way to estimate C^* from measurement of T_{50}. 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.

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.

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?

31. P. 6752, L3: It is not clear where the upper volatility bin limit comes from. This should be made clear. Also, it is not clear that the specific limit (C* >= 100 μg m^{-3}) is appropriate for an experiment where the mass loading is 650 μg m^{-3}. Ultimately, I find this discussion in this section to be a bit confusing, specifically with respect to how the various calculations were done.

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?

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.

34. P6754: Determination of volatility distributions from evaporation measurements (i.e. thermodenuder 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 γ = 1, then there is a specific volatility distribution that corresponds and should only be used if one continues to assume γ = 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.

35. P6754, L5: It should be noted that the inability to fit the α-pinene SOA mass thermogram with the model when γ > 0.01 is only true when the specific ΔH_{vap} distribution here is used. If the

ΔH_{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.

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).