I thank the reviewers for their detailed comments, which have helped to significantly improve the manuscript. I will address the individual comments below, indicating reviewer comments in italics.

Response to Referee #1

A very simple assumption of the temperature distribution has been made. In particular the radial gradient has been neglected. As the gas is heated/cooled via the tube wall this gradient is significant in the sections, where heating and cooling occurs. A justification for the assumption of a laminar flow (parabolic flow profile) should be given. In particular in the heating zone thermal turbulence could occur. The neglect ion of losses by thermophoresis should be justified/quantified.

As noted, we have assumed laminar flow in the thermodenuder (TD). Huffman et al. (2008) calculated that the Reynolds number in their TD (which has the same dimensions and flowrates as that considered here) was 38 at room temperature, thus well below the transition to turbulent conditions. Although the viscosity and flowrate change with temperature, the variation is not significant enough over the temperature range considered here to significantly influence this conclusion. This will be stated in the revised manuscript.

In our model we did not include losses by thermophoresis. Measurements by Huffman et al. (2008) indicate that increasing particle loss through the thermodenuder does occur as the temperature of the TD increases. The particle penetration decreased from ~95% at 25°C to ~80% at 230°C for NaCl particles. Because the model presented here explicitly determines the particle mass loss based only on evaporative loss it is possible to use an empirical correction that accounts for particle number loss to give a measure of the total mass loss (evaporation + particle loss). However, in experimental studies using thermodenuders it is common to either determine the mass loss based on particle size changes (which are approximately independent of particle number loss) or to correct for particle number losses when reporting final numbers. Therefore the neglect of particle number losses within the model is, to first order, consistent with experimental results.

Doing the model calculations with completely volatile particles is useful to demonstrate the features of the model. Adding an example for the more realistic case of particles, having a non-volatile core would be nice.

The inclusion of a non-volatile core will likely have a minimal influence on the results because the general evaporation behavior of the particles is governed by the thermodynamic properties of the volatile components. A non-volatile core provides only a secondary perturbation in that it changes the effective size of the particles (for a fixed volatile component mass), so long as the non-volatile core is assumed to be un-mixed with the volatile components (i.e. does not contribute to Raoult's Law vapor pressure depression). For example, if a non-volatile core were e.g. 50% of the total particle mass (assuming equivalent densities), the particle size would only increase by 26% for a fixed total organic mass and particle number. Thus, depending on the specific conditions (e.g. mass loading, C_{sat}, initial size) the calculated T_{50} would only increase by a few degrees, as is shown in Figure 7. We will explore this idea further and include a short discussion of our findings in the revised manuscript.

Response to Referee #2

Adding an appendix of model equations The governing equations of the model are two partial differential equations, which could be easily coded nowadays, e.g. by Matlab. Adding a compact list of model equations would help people understand and apply the model in their studies. Also it is necessary to explicitly write the initial and boundary conditions for the partial differential equations.

The equations are written explicitly within the text and all contained within a specific section (Section 2). Therefore it does not seem necessary to provide an additional table with these same equations. However, the few equations that are embedded within the text (i.e. not given as a numbered equation) will be given as numbered equations in order to make the overall equations governing the model more easily understood. Initial and boundary conditions will be given in the main text.

Moving sect 3.6 to 3.1, model validations should come before further discussions. The modeled results in Fig. 10 are steeper than the measurements. Since mono disperse aerosols in measurements are not perfectly mono-dispersed because of the DMA transfer function, the dispersion of observed spectra would be expected. I am not sure if this could explain the differences in Fig 10.

This section will be moved. Although some dispersion is to be expected due to particle size dispersion, given the relatively narrow transfer function of the DMA and the limited sensitivity of the TD model results to small changes in particle size (+/-20 nm, see Figure 7) this is unlikely to fully explain the differences in Fig. 10.

Adding tables for model configurations used in exemplary case studies (3.1-7)

This will be done in the revised manuscript.

Adding auxiliary files containing data of fig 1-10. Other people might benefit from these data when they try to quantitatively validate their coding of the model.

This will be done to some extent in the revised manuscript.

The Thermodynamics equation for the temperature (in the TD) is not solved in this model. Since prescribed temperature and velocity profiles are used in this model, the author should clarify that any kinds of profiles could be specified. Using measurement profile data (e.g. Faulhaber et al., 2009) could largely reduced uncertainties caused by the assumed profile introduced in the manuscript.

This will be clarified. It is true that using temperature profiles that are more specific to the TD in question could help to reduce uncertainties. The goal here was to use a temperature profile which generally conformed to the shape of various observed profiles (c.f. Huffman et al. (2008); Faulhaber et al. (2009)).

Since Td has been defined in the paper, it is clearer to use this notation, e.g p 2758, l 11: has been calculated at Td=Tref p 2771, figure: "temperature, Td : : :" p 2772, caption: ": : : at Td =25: : :"

This will be done.

Response to comment by Andrey Khlystov and colleagues

Khlystov's comment primarily centers on the consideration given to the role of using a denuder vs. not having a denuder. In the presented analysis, it was assumed that when a denuder was not present loss to the walls was negligible and thus recondensation within the cooling section was determined to be an important issue for certain vapor pressure/aerosol loading conditions. (Note that for some conditions, notably those most directly applicable to ambient measurements, recondensation was not found to strongly influence the results regardless of whether a denuder was or was not present in the simulation). Khlystov argues that the assumed boundary conditions for the no-denuder case was incorrect, pointing out that having a tube that is at lower temperature than the heated section will lead to condensation of gas-phase species onto the tube, i.e. that the room-temperature tube effectively behaves as a denuder. It was also suggested that as the walls of the tube become completely coated with the compound(s) of interest (presumably after some start-up and equilibration time) the correct boundary condition becomes one where the gas-phase layer adjacent to the tube walls should be considered in equilibrium with the walls at the saturation vapor pressure of the compound(s) of interest. As such, Khlystov argues that our imposed boundary condition overestimates the likely magnitude of the (potential) recondensation bias. It is specifically argued that "*This boundary condition is critical because it is what leads the aerosol-vapor system to equilibrate, contrary to the conclusion of the author. Further, this also illustrates why a denuder should not be incorporated in the cooling section of a TD employed to estimate aerosol thermodynamic properties; the denuder will prevent the system from equilibrating at the cooling section temperature, and thus provides a poorly characterized endpoint for inferring thermodynamic properties.*" First, it must be noted that the suggestion by Khlystov that the boundary condition is the saturation vapor pressure is also an assumption. It is mentioned that he has observed the formation of coatings of dicarboxylic acids on tubing walls downstream of the TD heated section, but this does not definitively demonstrate that the boundary condition should be C_{sat} for all cases, only that there is some capacity for uptake of gas-phase molecules by the walls of tubing. Nonetheless, the assumption of equilibrium with the walls is reasonable and we consider the implications further below.

We have modified our "no-denuder" model to have as a boundary condition at the walls of the cooling tube the saturation vapor pressure, as suggested by Khlystov. Specifically, we assume that there is an infinitesimally thin region near the walls in equilibrium with the walls (assumed to be saturated with the compound(s) of interest) in the cooling section and that the capacity of the walls to absorb gas-phase material is infinite. Compared to the previously considered denuder case, this has the effect of decreasing the rate of mass transfer to the walls because the driving force (the concentration gradient) is lessened and also provides a limit on the total amount of material that will condense to the walls. Indeed, recondensation is lessened compared to the non-absorbing condition, although our model indicates that some recondensation does still take place, particularly in the bins closest to the center of the tube (i.e. furthest from the walls). However, it should be noted that even the model with the denuder occasionally shows some recondensation for particles in the center of the tube (depending on the exact conditions) so this result is not particularly surprising. In the revised manuscript, results will be presented using the updated boundary conditions for the non-denuder cooling section. However, in the revised manuscript we will also present results using our initially assumed boundary condition (i.e. no uptake) as this provides an upper-limit on the potential role of recondensation. It will be made clear that this is indeed an upper limit that may not actually occur in practice, especially if the residence time in the cooling/transfer tubing is kept short (see discussion below).

Khlystov rightly points out that the presence of the denuder section can lead to a decrease (negative bias) in the mass fraction remaining (MFR), while recondensation leads to an increase (positive bias) (with bias being defined as a deviation from the MFR that exists at the exit of the heated section). This is apparent in the presented calculations. While reconsideration of the boundary conditions in the cooling section as per Khlystov's suggestion leads to a decrease in the calculated contribution of recondensation, it is important to recognize that there are numerous groups who are now making use of thermodenuders that explicitly include a denuder/cooling section and it is therefore important that the entire system (heated section *and* denuder) be understood to allow for robust interpretation of such observations. In this context, whether the denuder imparts a bias to the measurements or not is incidental: denuders are in use and must therefore be accounted for and a physically realistic model employed. Khlystov makes the case that the inclusion of the denuder section is unnecessary and perhaps even undesirable, but this does not change the fact that such systems have been in use and will (likely) continue to be used in the future.

On p.2760 the author states that "When the residence time in the heated section is increased, it is also increased by a proportional amount in the denuder section." This asserted proportionality is arbitrary, and the ensuing analysis is highly misleading. In reality, one may design the cooling section as needed. Additionally, evidence is offered by Khlystov that no significant recondensation is observed using a "more complete" model (Saleh, 2008).

For a fixed system this proportionality is not arbitrary. However, Khlystov is correct that a given system can be modified to have a shorter cooling section (whether this be a tube or a denuder) and the revised manuscript will make this point clear, specifically the point that the residence time following the heated section can be made quite short if the system is designed as such. In fact, if our model is modified to have the same dimensions and residence times as in the Saleh study (and using the vapor pressure and enthalpy of vaporization for adipic acid with T_d = 45 °C) it is similarly found that recondensation is not significant. However, it should be noted that this conclusions is independent of whether the saturation boundary condition or no-uptake boundary condition is used.

On p. 2761 and the related statement in the conclusions about the equilibration times: "Low volatility compounds are not all that sensitive to the denuder section, however they will not reach equilibrium in the heated section in any reasonable (i.e. experimentally accessible) time period." The author gives no justification for the second part of the statement. The statement and the corresponding conclusions about the equilibration times are highly misleading because they are not qualified by the specific conditions modeled by the author, and thus falsely give an impression of universality. In reality, the equilibration time depends not only on the C_{sat}*, but also on the concentration of aerosol and its size distribution, which is clear from Eq. 1.*

Khlystov rightly points out that whether "low volatility" compounds (here, I will take this to mean compounds with C_{sat} values <= 1 μg/m^3) reach equilibrium in a "reasonable time period" (here, < 60 seconds residence time) in the heated section does depend on more than just the compound vapor pressure and that, in particular, aerosol concentration is of particular importance. In the revised version, we will make this statement more precise, specifically discussing that this is true at low loadings (ca. < 100 μg/m^3 but how at higher mass loadings equilibrium may actually be reached. This is important as it indicates that there are different considerations that must be taken into account for analysis of ambient (typically low loadings) vs. laboratory (often done using high loadings) measurements. This discussion will be supported with a figure such as the following, where C_{sat} = 1 μg/m^3 was used and the MFR at the end of the heated section (i.e. prior to cooling) is shown for different mass loadings, assuming the walls in the heated section to be non-absorbing:

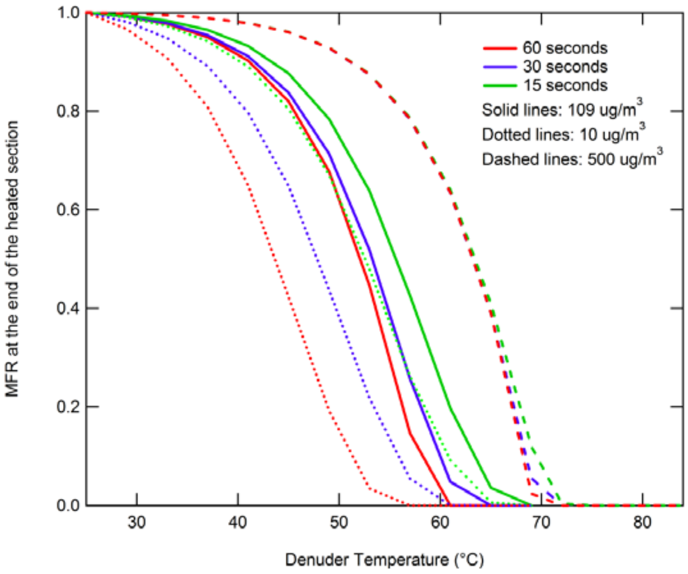

Figure 1. The calculated mass fraction remaining at the end of the heated section is shown as a function of the thermal denuder temperature for a single compound with C_{sat} = 1 μg/m^3 for different C_{OA} values and different residence times (see legend for specific values).

The statement in the conclusions "Unlike previous models, separate heating and denuder sections are explicitly accounted for as is the velocity profile through the denuder tube" exaggerates the achievements of the presented model. We have previously described a size-resolved model for aerosol evolution in wet annular and flat denuders that takes into account velocity profile, hygroscopic growth and transfer of the gas species to and from the particles and the walls (Khlystov et al., 2009). An adaptation of this model to thermodenuders, which includes heat exchange with the walls, was used to test the effect of re-condensation and an absorber in the cooling section (Saleh et al., 2008). Unlike our model, the one presented in this manuscript assumes that the radial temperature profile is constant (p.2759 line 29) and works only with monodisperse aerosol. The size distribution of particles may have a profound effect on the evaporation/condensation rates because of the strong dependence of evaporation rate on particle size. Radial temperature gradients are extremely important too. In the transition regions (cold to hot and hot to cold) the temperature gradient is the leading cause of different evaporation/condensation rates experienced by particles at different distances from the wall. These *differences are critical for the evolution of the particle sizes and gas phase concentrations in the system. Unfortunately, these radial gradient effects in the transition region are ignored in the model presented in this manuscript (the temperature is assumed to linearly increase or decrease with axial distance, while being uniform in the radial direction). Since the presented model does not take into account radial heat transfer, the only source of radial concentration gradient in the heated section is due to the differences in the time particles spent there. The author does not indicate how strong these radial gaseous concentration gradients are and whether they are significantly larger than the axial diffusion (the equations indicate an implicit assumption that the axial diffusion is negligible).*

The previous work of Khlystov and colleagues is noted and will be clearly referenced and their contributions acknowledged in the revised manuscript, and the statement in question will be revised. Axial diffusion is assumed negligible in this model, similar to the assumption made by Saleh and Shihadeh (2007). This will be made clear. We will also make clear the limitation of our model with respect to the neglect of radial temperature gradients. Specifically, it is understood that, for the heat-up section, particles closest to the wall (where the residence time is longest) will experience a temperature that is somewhat greater than the mean temperature and thus these particles will evaporate to a greater extent. However, during the cool-down section the particles closest to the wall will experience a temperature that is somewhat less than the mean temperature and thus these particles will evaporate to a lesser extent.

The conclusion that the denuder is most important for high loadings is also questionable. It is based on the larger deviation between the "on" and "off" cases at higher concentrations. First of all, as discussed above, the MFR for the "off" case are most probably grossly overestimated, because the condensation to the walls was erroneously switched off. Secondly, no proof is given that the negative bias due to the gas stripping by the denuder would not increase at higher aerosol concentrations. In fact, Figure 5 shows that for the "on" case most trajectories demonstrate a significant decrease in particle size, such that at the end of the denuder the particle size is well below the size at the end of the heated section. This must lead to a strong negative bias in the "on" case. The conclusion about the importance of the denuder at high C_{OA} *appears to be the exact opposite to what the model indicates: the denuder introduces a larger bias at higher* C_{OA} *than at the low* C_{OA}*. Granted, the author acknowledges that Figure 5 is in effect* *comparing apples to oranges: a high C_{OA} / high C_{sat} compound vs. low C_{OA} / low C_{sat} one, i.e. there are two competing factors in play: C_{OA} and C_{sat}. The only reason why the author picks C_{OA} over C_{sat} as the most important factor in re-condensation is: "a larger set of runs (not shown) indicates that C_{OA} plays a more important role than C_{sat} in determining the influence of re-condensation." It is not clear why the data supporting this is not shown, while the negative bias due to the denuder is glossed over.*

A subset of the full set of calculations was shown and, as is often done, representative calculations were shown. Also, a physically plausible reason for understanding the conclusion is advanced: "At lower loadings, there is less total particle surface area available for re-condensation, which has the effect of slowing the condensation." (p. 2759, line 11) The focus of the discussion, as presented, was on the influence of re-condensation, and when considering only re-condensation (at least the upper limit determined from consideration of the non-absorbing non-denuder), we do find that C_{OA} is the more important parameter than C_{sat}. However, as pointed out, there can certainly be a strong influence of the denuder leading to additional shrinking of the particles; this was explicitly acknowledged with the discussion relating to Figure 3 and so it is not clear in what context this has been "glossed over". However, in the revised manuscript we will make clearer this point in the context of the temperature dependent simulations as well and make a stronger connection between the results presented in Figure 3 (evaporation in the denuder) and the temperature dependent results. Specifically, we will discuss that, in the context of continued evaporation in the denuder, C_{sat} is the more important parameter than C_{OA}. We will also add results from the model when the saturation boundary condition suggested by Khlystov is used.

It is also worth mentioning here that there are clearly different considerations between very high ($C_{OA} >$ 1 mg/m^3) vs. comparably low ($C_{OA} <$ 100 μg/m^3) with respect to the importance of re-condensation. For such very high loadings, significant re-condensation is predicted from the model independent of the boundary condition imposed in the cooling section for compounds both with relatively low saturation vapor pressures ($C_{sat} <$ 1 μg/m^3) and relatively high vapor pressures ($C_{sat} >$ 100 μg/m^3). This is because at such high loadings the particle surface area becomes large compared to the wall surface area. However, it should be noted that the higher vapor compounds are still calculated to shrink to some extent within the denuder as there is still a competition between re-condensation and loss of gas-phase species to the walls (as discussed above). Also, if the cooling section is made extremely short (as suggested by Khlystov) it may be possible to minimize re-condensation, although it then becomes important to ensure that re-condensation does not take place after the cooling section (i.e. in any further transfer tubes or within instrumentation), or that this potential further re-condensation is accounted for in any analysis.

Figure 4 shows MFR (mass fraction remaining) at different mass loadings and is used by the author to conclude that the use of an absorber is very important at high aerosol concentrations (p.2759 "Figure 4 shows that the presence of the denuder section is most important for high loadings, which are typical of laboratory experiments."). Figure 4, however, only compares the cases when the denuder is used or not in the cooling section (the "on" and "off" cases, respectively). The "off" case is most probably biased high due to the re-condensation. The "on" case is assumed to be correct or to have a smaller error, which is probably not the case. In fact, Figure 3 demonstrates that the denuder can absorb more than 50 g/m3 of material causing a strong negative bias for semi-volatile aerosols even at room temperature. At higher gas concentrations originating from the heated section this bias will be even larger, due to a stronger driving force (concentration gradient in the first term of Eq.1). The magnitude of this negative bias is not shown in Figure 4. Therefore, the conclusion about the importance of the denuder at any aerosol concentration is not supported by the figure and is, actually, wrong.

The labels on Figure 4 were unfortunately imprecise. The solid lines refered to the MFR after the aerosol has passed only through the heated section while the dashed lines are the MFR after the aerosol had passed through the both the heated section and the cooling section (i.e. the denuder with the denuder "off"). By comparing these curves this figure presents a measure of the influence of re-condensation on the MFR in the absence of the denuder (under the assumption that there is no loss to the walls) and it is clear that at the higher loading (for C_{sat} = 10 μg/m^3) the influence is greater. To the extent that this figure addresses the influence of re-condensation in the absence of loss to the walls it is correct (albeit with improper labels). However, Khlystov is also correct that with the denuder in the "on" position the particles can actually shrink somewhat due to losses of the gas-phase in the denuder section in comparison to the particles that had passed only through the heated section (i.e. would have a "negative bias"). This figure will be fixed in the revised manuscript.

Finally, Khlystov raises an important question regarding mass balance in the model (p. C214), specifically asking "*The question is: since there is no absorption by the walls in the heated section, where does this* *evaporated material go?*". It has been verified that mass is conserved in the heated section by calculating the total organic mass (particle + gas) at all points in the simulation and ensuring that this is constant throughout the heated section (note that mass can be lost in the denuder section when the denuder is "on"). Particles closest to the wall can evaporate to a greater extent than particles near the center of the tube because there is a radial concentration gradient established in the heated section as the particles closest to the walls evaporate to a greater extent than those in the center of the tube for a fixed distance traveled (as a result of the particles near the wall having a longer residence time). Gas phase molecules therefore diffuse from high (near wall) to low (center of tube) concentration, which allows the particles nearest the wall to continue to evaporate. We have also calculated the expected size decrease if the particles were to reach equilibrium at the TD temperature using the equation given by Khlystov: $r = r_0(1-dC/C_{OA})^{1/3}$, where r is particle radius, dC is the amount of material needed to saturate the air at the new temperature ($dC = C_{sat}(T_h) - C_{sat}(T_0)$, where T_h is the TD temperature, T_0 is the entrance temperature and C_{sat} is the saturation concentration at a given temperature) and C_0 is the initial aerosol mass concentration. Khlystov gives dC = 53 μg/m^3; however, I calculate that dC actually is 112 μg/m^3 for the high C_{OA} case shown in Figure 5. This comes from C_{sat}(25°C) = 100 μg/m^3 and C_{sat}(35°C) = 212 μg/m^3 (assuming ΔH_{vap} = 60 kJ/mol), which gives dC = 112 μg/m^3. Therefore, the equilibrium size would be 63.3 nm (for C_{OA} = 150 μg/m^3) instead of the 86.5 nm given by Khlystov. Thus, only the particles in the few bins nearest to the wall shrink beyond their apparent equilibrium size, and this occurs for the reason discussed above. For the low C_{OA} case (C_{OA} = 15 μg/m^3; C_{sat} = 1 μg/m^3; ΔH_{vap} = 120 kJ/mol; T_h = 60°C) dC = 145 μg/m^3 – 1 μg/m^3 = 144 μg/m^3, and therefore given sufficient time the particles would evaporate entirely, i.e. the system would never reach saturation. This will be the case whenever $C_{sat}(T_h) - C_{sat}(T_0) > C_{OA}$.

To summarize, Khlystov raises a number of important points, with most of the concern focused on the assumption that in a system with no denuder (but a tube of equivalent length) molecules are not adsorbed (or absorbed) to the walls. This assumption provides an upper limit on the possible influence of recondensation, and is therefore an interesting limit to consider, but as Khlystov notes this may not be the most physically plausible boundary condition. In the revised manuscript, the boundary condition suggested by Khlystov will be explicitly considered. It should be noted that this discussion has no influence on the conclusions with respect to the influence of the denuder. However, in the revised manuscript it will be made clearer that the presence of the denuder can introduce a "negative bias" with respect to the state of the aerosol when it leaves the heated section in the thermodenuder.