General Comments

This paper report interesting results on how the thermally-induced evaporation of SOA particles depend on the mass-loading. The paper is well written and structured. I mainly have some criticism toward a few of the model assumptions. This primarily concerns the assumed initial gas and particle composition at the inlet to the thermodenuder (TD). Since you only have a residence time of ~1 minute in the flow-tube I don't think it is correct to assume (i) that the SOA particles are in equilibrium with the gas-phase at the inlet of the TD at low mass loadings (low condensation sink) and (ii) that monomer-dimer equilibrium is reached within this time scale. I am also a bit skeptical to the way you first calculate the monomer gas/particle equilibrium distribution and then after this calculate the monomer/dimer equilibrium distribution (see specific comment below).
When you have addressed these points preferentially with additional model simulations of the non-equilibrium SOA formation in the flow-tube and carefully answering my specific comments below I think the paper is suitable for publication in ACP.

P 10001, L1-4:
"In addition, several experiments have observed slower than expected room-temperature evaporation of both ambient (Vaden et al., 2011) and laboratory generated (Saleh et al., 2013; Grieshop et al., 2007; Wilson et al., 2015) SOA during isothermal dilution."

Vaden et al., 2011 also studied laboratory generated SOA. Refer to Vaden et al., 2011 for the laboratory generated SOA too.

P 10002, L3:
You use the term *"homogeneous nucleation"* to refer to how the SOA particles were generated. I have also used this expression in previous publications when I refer to new particle formation during no-seed SOA particle formation experiments. However, I don't know if this is correct. Lately I have started to use "formation of nano condensation nucleii (nano-CN)" instead, with a reference to McMurry P. H., Kulmala, M., Worsnop D. R.: Special Issue on Aerosol Measurements in the 1 nm Range, Aerosol Sci. Technol. 45, i, 2011.

P 10005 L1-L4: *"Here, to provide for more consistent fitting and since no evaporation at room temperature was observed, the fit curves were forced to go through unity at room temperature."*

Change to: "Here, to provide for more consistent fitting and since no evaporation was observed at room temperature, the fit curves were forced to go through unity at room temperature."

Sect. 2.4.1 Thermodenuder model.

An assumption that I think should be mentioned is that you assume that the particles behave as liquid droplets (no mass transport limitations in the particle-phase). This may be justified by the relative high temperature in the TD but is probably not entirely true for room-temperature isothermal evaporation.

P 10005, L12-13: *"it is assumed that the gas/particle system is at equilibrium before entering the TD."*

For low mass-loadings the initial equilibrium gas-particle may not be valid because the residence time in the flow tube was only approximately 1 minute? How would this influence the results? Also to assume monomer-dimer equilibrium at the entrance into the TD may not be correct if $1/k_r$(298 K) is not much larger than the residence time in the flow tube.

P 10006, L14-21: *"If K_{eqm} is large then all condensed-phase species would be in dimer form and, at equilibrium, all gas-phase material would be drawn into the condensed phase. Here, this situation is avoided through the following simplification to determine the initial particle state at the TD inlet. First, the gas/particle (monomer only) equilibrium distribution is calculated given the specified volatility distribution and C_{OA}. Then the monomer/dimer equilibrium in the condensed phase is calculated, and the gas-phase concentrations are set to zero to avoid large amounts of condensing material at the next time step. Since a charcoal denuder is placed immediately after the flowtube, this simplification is physically accurate."*

It is good that you clearly describe the assumptions that you use for the model setup but I think it would be more physically reasonable to assume that monomers are not dissolved (absorbing) into the dimer SOA volume fraction if you want to limit the growth during next time step. You would then have to simulate (or iteratively derive) the SOA composition at the TD inlet. To assume that the monomers absorbs into the total C_{OA} would not be correct then. Especially for the low SOA loading experiments, I think that you actually need to explicitly simulate how the non-equilibrium SOA formation and dimer/monomer SOA composition changes in the flow-tube. It would also be good to simulate the vapor stripping in the charcoal denuder and not just assume perfect gas-phase removal. Do you have some experimental results to justify this assumption?

P 10007, L3-4: *"The rate at which dimers decompose is governed by k_r and k_f, both of which are likely to be temperature dependent."*

To me it is not entirely clear if you always assume dimer/monomer equilibrium in the model or if you explicitly simulates the non-equilibrium dimer and monomer composition and how it changes in the TD as a function of temperature and evaporation. You need to explain this more clearly.

P 10009, L23-24: *"Regardless, it is apparent that the effective volatility of the SOA at C_{OA} is not higher than at low C_{OA} and that, despite the slight differences, the response to heating"*

Add "high"

"Regardless, it is apparent that the effective volatility of the SOA at high COA is not higher than at low COA and that, despite the slight differences, the response to heating"

It is interesting to see that the thermograms do not either seem to be influenced by the initial particle size (dp,V,bypass). To me this is an additional indication that the particle evaporation was not driven by the volatility of the particles but the decomposition rate of dimers (oligomers) back to volatile monomers (see Roldin et al. (2014)).

P 10013, L20-24: *"At smaller K_{eqm}, extensive room temperature evaporation occurred as a result of the increasing initial fraction of semi-volatile monomers, a result that is inconsistent with the observations. However, even for the simulations at larger K_{eqm}, some evaporation at room temperature was always predicted."*

Yes but this partly because you assume liquid SOA particles. If the SOA particles are solid-like at room temperature (as suggested by several studies), the evaporation of monomers would slow down substantially once the particle surface layer has been filled with non-volatile dimers.

P10015, L15-18: *"The range of k_r independently determined here are somewhat larger than the room-temperature k_r suggested by Trump and Donahue (2014) (= $1.1x10^{-4}\ s^{-1}$) and Roldin et al. (2014) (= $2.8x10^{-5}\ s^{-1}$), which were based on needing an evaporation timescale of 1 h for isothermal evaporation (Grieshop et al., 2007; Vaden et al., 2011). However, their estimates may not have fully accounted for the dynamic nature of the system, and thus underestimated the actual dimer decomposition rates compared to that obtained here."*

It is true that we used k_r=2.8x10^{-5} s^{-1} for the results presented in Fig. 6 in Roldin et al. (2014) but we also tested other values of k_r. Including k_r=12 h^{-1} (0.0033 s^{-1}) for a group of relatively abundant (~20 mass %) and shout-lived dimers, in combination with more long-lived but less abundant (1-2 mass %) dimers with k_r=1/30 h^{-1}. We were then able to accurately simulate the nearly particle size independent evaporation of fresh SOA particles from the experiments in Vaden et al. (2011) (Fig. 7 and Fig S9-S10 in Roldin et al. (2014)). For these simulations we considered that the particles had a high viscosity in agreement with Abramson et al. (2013). However, with this setup we substantially overestimated the effect of particle ageing in the Teflon chamber on the observed evaporation rates. This can be an indication that the actual oligomer (dimer) fraction of the short-lived dimers was larger than 20 % (maybe close to 100 % as you suggest). This would have limited the effect that VOC wall losses had on the particle composition (and evaporation behavior) when they were aged in the Teflon chamber by Vaden et al. (2011). For these type of experiments I generally think that it is important to also explicitly simulate the SOA formation phase and not just the evaporation stage of the experiments because if you don't get the model to agree with the observations both for the formation and the evaporation experiments something is not correct in the model.

P10017, L4-10: *"The simulated MFR values at the end of 15 h of SOA evaporation are somewhat lower than was observed in the literature experiments for dry, fresh SOA from α-pinene + O_3, where MFR ~0.35–0.4 at 15 h (Vaden et al., 2011; Wilson et al., 2015). However, the extent of evaporation is dependent on the model assumptions, specifically the k_{loss} and DF. Smaller k_{loss} or DF leads to larger MFR at a given time due to more extensive inhibition of evaporation resulting from faster* *saturation of the gas phase (Fig. 7a). As neither the k_{loss} nor DF are explicitly known for the literature experiments, a more quantitative comparison is not possible. Conversely, larger k_{loss} or DF leads to more extensive evaporation."*

The effect of k_{loss} and DF on the particle evaporation is an important finding that need to be addressed in future isothermal evaporation experiments. The Vaden et al., (2011) evaporation experiments were performed in a very small (7 L) stainless steel chamber with active charcoal on the bottom of the chamber. Before the SOA particles were introduced to the evaporation chamber they passed two charcoal denuders. In Roldin et al. (2014) we evaluated the VOC losses to the charcoal by simulating the DOP experiments from Vaden et al. (2014). Based on this we could conclude that the gas-phase uptake to the charcoal denuder was relatively efficient. We never reported the value of k_{loss} but it was on the same order of magnitude that you use (4×10^{-3} s^{-1}). However, the gas-phase VOC concentration is also governed by the SOA particle concentration in the evaporation chamber. In the Vaden et al. (2011) experiment the total particle concentration was very low ~100 cm^{-3} (~1 μg m^{-3}). Thus, the gas-phase VOC concentration became very low although the uptake to the chamber walls (charcoal) was not instantaneous.

P10017, L16-21: *"Simulations using the dimer-decomposition model with different starting particle sizes show some dependence on particle size (dp = 90, 180 and 360 nm), with larger particles having smaller MFRs at a given time (Fig. 7a). However, the overall differences are relatively small and reasonably consistent with the observations given that the observations have typically considered a narrower size range than examined here."*

I still think that the differences between the different particle sizes in Fig 7a is relatively large and it shows that something is missing in the model in order to explain the nearly size independent evaporation rates reported by e.g. Vaden et al. (2011). As mentioned previously several studies (e.g. Virtanen et al., 2010; Abramson et al., 2013 and Zhou et al., 2013) have shown that SOA particles are not liquid-like but very viscous tar or even solid-like. I think it would be appropriate to mention that the mass-transfer limited diffusion within the particle-phase will also influence the isothermal evaporation but that this was not considered. What was the RH in the flow tube?

P10020, L13-16: *"If the particles were primarily semi-volatile monomers for which evaporation were limited by diffusion in the particle phase, then changes in viscosity should lead to substantial increases in the observed evaporation rate (Zaveri et al., 2014)."*

Do you mean:

If the particles were primarily semi-volatile monomers for which evaporation were limited by diffusion in the particle phase, then decreasing viscosity should lead to substantial increases in the observed evaporation rate (Zaveri et al., 2014)."

This we also showed in Roldin et al. (2014) (Fig 5c).

P10022, L19-22: *"Thus, it seems that a hybrid model where the particles are* *composed of a substantial fraction of dimers (or oligomers) and some smaller fraction of low-volatility compounds may ultimately provide a more complete description."*

I fully agree.

I suggest that you add the evaporation curves from Vaden et al. (2011) to Fig. 4b and Fig. 7.