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# *Interactive comment on* "The influence of particle composition upon the evolution of urban ultrafine diesel particles on the neighbourhood scale" *by* Irina Nikolova et al.

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Please see the attached zip file, which contains a formatted version of the reply to the referees, a revised manuscript version with tracked changes, and a clean version of the revised manuscript.

REVIEWER #1. The paper presents a targeted model study on the evaporative shrinking of diesel particles over timescales representative for the dispersion on the neighbourhood scale. The model study explores the sensitivity of nucleation-mode diesel particles to the volatility of the initial chemical composition represented by a mixture of surrogate molecules, the higher n-alkanes, in a very systematic manner. Gaussian

distributions of the n-alkanes with carbon numbers between 16 and 32 with various bandwidths are systematically tested for three different vapour pressure datasets. The influence of a non-volatile core with various mass fraction in the nucleation mode is also tested. A new concept of threshold modal composition is introduced which allows identification of the components for which an accurate vapour pressure estimation is most critical. The paper is well written for the most part and results are presented in a structured way. Input datasets and model results of the study are well documented and easily accessible. Answer: we thank the referee for their assessment of the scope and importance of the manuscript.

It is claimed that the method has the potential to improve the efficiency of urban aerosol models. I had expected to see a comparison of the suggested simplifications with the full model and an estimate of the computational savings. Answer: such a comparison is beyond the scope of the paper. We have removed the final sentence in the abstract.

The main problem is that the analysis was restricted to the process of condensation/evaporation alone. Answer: CiTTyCAT UFP contains treatments of coagulation, deposition, and exchange with the free atmosphere. Our earlier work (Nikolova et al., 2016, Faraday Discuss.,189, 529-546, DOI: 10.1039/C5FD00164A) has shown that deposition and coagulation have a minor effect in the current scenario and so were switched off to allow a more straightforward diagnosis of model behaviour. We have amended the revised ms at line 208-210 of the version with tracked changes (see manuscript version appended) to say: "Our earlier work (Nikolova et al., 2016) has shown that deposition and coagulation have a minor effect in the current scenario and so were switched off to allow a more straightforward diagnosis of model behaviour."

Specific Comments 1.) The exhaust size distribution from laboratory test-rig measurements (Figure 2-S) shows a broad distribution of n-alkanes in the diameter range < 10-18 nm with a maximum at $C_{29}H_{60}$ and a range of +/-7 corresponding to sigma = 3. In contrast, most realisations of the current model setup resulted in solutions with a narrow distribution of n-alkanes with carbon number < 18 (this includes all solutions

for vapour pressure datasets A-a and B-c ) to explain the REPARTEE-like behaviour. These n-alkanes are predominantly in the gas-phase under ambient atmospheric conditions. Only for vapour pressure dataset B-c, which has the highest vapour pressure for all n-alkanes, the model gives solutions with carbon number > 20 and broad distributions (sigma = 3 to 5) for the REPARTEE-like behaviour (P.15, lines 372-373). It looks like other processes, consistent with an initial nucleation mode composition of higher-carbon-number compounds, are relevant. Answer: In this study we propose a systematic method to evaluate the evaporative potential of nucleation mode particles (diameter less than 30 nm) of varying composition under ambient conditions. Our approach is to 'scan' the available compositions efficiently using the 'mode composition plus sigma' method we describe, and then âĂŤ subsequently âĂŤ to compare model outcomes to the field observations and the laboratory measurements. That is, we do not assume a priori that any given vapour pressure set or composition is correct. We do not need any other aerosol processes in the model to find compositions that are consistent with the field observations and lab experiments.

2.) Coagulation was obviously not included in the model setup (P.8, line 192). Coagulation is enhanced through the shrinking of particles by evaporation (Jacobson et al.,2005). As particles shrink, their coagulation rates with larger particles will increase. Although evaporation has a greater effect on the evolution on the particle size distribution in the first second, coagulation will become more important towards longer time scales and may be dominant at 100 s. When evaporation and coagulation are treated together, there will be a feedback of coagulation on the composition of nucleation mode particles. This is because coagulation affects the vapour pressures of n-alkanes in particles of a given size through the Raoult's Law. This feedback may well influence the calculated nucleation mode peak diameter. Answer: Coagulation was not included in the model setup for this scenario, as discussed above. Our coagulation tests used the same model and so included the size-dependence of coagulation rates. Text amended as described above.

3.) P.5, lines 114-116: The range of volatility of the particle composition in the diesel exhaust emissions is not known. However, formation of a volatile nucleation mode is evident in a laboratory system that mimics real-world dilution of diesel exhaust (Arnold et al., 2012; Rönkkö et al., 2013). Aerosol dynamics models show that low volatile and extremely low volatile organic compounds are required to explain the evolution of the volatile nucleation mode during cooling and expansion of the exhaust (Rönkkö et al.,2013; Pirjola et al., 2015). Answer: Our study does not consider the cooling and expansion of the exhaust plume. Instead, we have examined the evaporative potential of particles under ambient conditions, timescales ~100 s, distances 10 m - 1 km and size distribution that is typically observed in proximity to traffic sites (Nucleation mode Dp = 23-25 nm, Aitken mode Dp at around 50-60 nm, Dal'Osto et al 2011). The REPARTEE-like behaviour could only be explained if low-carbon-number compounds are present in the particle. However, the evaporation is dependent on the vapour pressure and we show in Figure 6-S the relative differences in the nucleation mode peak diameter using B-c (highest vapour pressure values) and A-a (lowest vapour pressure values). Overall, the largest differences in the 100-s $D_{pg,nuc}$ occur for modal compositions between $C_{22}H_{46}$ and $C_{24}H_{50}$ and composition standard deviation from 1 to 3. i.e., the range where the highest relative differences occur is not in the low-volatile nor extremely-low volatile organic compounds. We have amended the text at line 138-139 to remind the reader that the simulation is focused on events after dilution and cooling, in section Introduction, as follows: 'The model simulations are focused on events after dilution and cooling of the exhaust-pipe plume.'

4.) P.8, line 184: A higher number of bins representing the small particles would give a more accurate result for the relevant part of the size distribution. How does a larger number of size bins, for instance 25, change the result on the REPARTEE-like behaviour? Answer: In our study we use 15 size bins, 6 of which are located in the nucleation mode. This provides sufficient details in the nucleation mode and a reasonable amount of size bins, attractive for implementation in large 3-D models. We cannot see a reason in the physics for a substantive change in our results when running with

more bin resolution.

5.) P.9, line 197: how fast is the dilution in the first second? Preferably, the dilution ratio of the modelled total particle number after 1 s should be given and compared to literature data. Answer: The model simulations are focused on events after tailpipe dilution and cooling. Dilution has been considered for gas-phase concentration of the participating compounds in the box. Background gas-phase concentration is kept at zero. This information is now added in lines 281-282.

6.) P.11, line 251: Gas-phase n-alkanes $C_{16}H_{34}$ and $C_{17}H_{36}$ can be oxidized by the OH radical leading to oxygenated products with lower vapour pressures (Jordan et al., 2008). An estimate should be given how much gas-phase oxidation can affect the modelled size distribution on the time scale of 100 s. Answer: The rate coefficient of $C_{16}H_{34}$ according to Atkinson and Array (2003) is $23.2\times10^{-12}$ cm3 molec-1 s-1. The hydroxyl (OH) radical concentration is typically around $10^6$ molec cm-3. Hence the timescale for oxidation of $C_{16}H_{34}$ is about $10^6$ s, a time much larger than the 100 s considered in this study. We add at line 279-281: "For hydroxyl (OH) radical concentration $\sim10^6$ molec cm-3, the timescale for oxidation of $C_{16}H_{34}$ is about $10^6$ s (Atkinson and Arey, 2003). Therefore oxidation of SVOC is neglected given the timescale in our study (100 s)."

7.) P.17, lines 409-410. Which assumptions were made regarding the mixing with the non-volatile core? If the non-volatile component is able to influence the gas-particle partitioning, for example due to its absorptive nature, then the evaporation rate will be affected by a decrease of the vapour pressure of the volatile compounds in the mixture through the Raoult's Law. If the non-volatile core is not in a mixture with the n-alkanes, there is no Raoult's Law depression of the vapour pressure of the volatile compound. The assumption about the non-volatile core will also affect the conclusion about its chemical composition in the nucleation mode (organic carbon or lubricant oil metal compounds). Answer: In this study, the non-volatile component is not considered as part of the mixture, therefore does not affect the saturation vapour pressure of the

volatile compounds. The activity coefficient in our study is assumed to be unity in the absence of any additional information about the ideality of the solution. In-particle-phase processes and in-particle chemistry are not considered. The referee is correct to point out the difference effects that organic carbon and metal compounds will have on SVOC vapour pressures and we have corrected the ms so that it now reads at line 208-210: "The condensation/evaporation process applies Raoult's Law (for an ideal solution of the volatile compounds) and a mass accommodation coefficient $\alpha = 1$ (Julin et al., 2014) for all SVOC"

Technical Corrections Abstract p.3, line 52-54. Mention the uncertainty range of the vapour pressures for n-alkanes between C22H46 and C24H50 based on the vapour pressure datasets displayed in Figure 3. Answer: We have added information on the uncertainty range as suggested as follows (lines 260-261): 'The vapour pressure ranges of C22H46 and C24H50 are between [9.23x10-3 and 8.94x10-6 Pa] and [2.26x10-3 and 2.46x10-7 Pa], respectively.'

P.4, line 71: Add information on size range. Answer: Information on the size is added at line 76ff, as follows: 'Harrison et al. (2011) reported that on a busy highway in central London, UK, 71.9% of particles by number were traffic-generated; of which 27.4% are found in the semi-volatile exhaust nucleation mode (size between 15 and 30 nm), 38% are in the exhaust solid mode (size > 30 nm) and the remaining 6.5% are from brake dust and resuspension (size > 2000 nm).

P.4, line 79: Jacobson et al. (2005) and Karl et al. (2016) should be included in this list. Answer: The references are included at lines 82ff, as follows: 'Experimental and modelling studies have advanced our understanding of the behaviour of urban air UFP, e.g. the relevant aerosol dynamics important to the evolution of the UFP in space and time (Jacobson, 2005; Allen et al., 2007; Biswas et al., 2007; Dall'Osto et al., 2011; Nikolova et al., 2011; Karnezi et al, 2014, Karl et al., 2016).'

P.7, line 168: missing ", respectively" at end of this line. Answer: Done.

P.10, line 227-232 and Figure 3: the vapour pressure data from Lemmon and Good-win(2000) should be included in Figure 3 to facilitate comparison with previous studies on the volatility of exhaust particles near roadways (Zhang and Wexler, 2004; Zhang et al., 2004; Karl et al., 2016). Answer: Figure 3 and caption are now updated with Lemmon and Goodwind (2000) vapour pressure, which has a range of vapour pressure is very similar to Compernolle et al. (2011). Also, we have added the reference in the description of vapour pressures parameterisations in section 2.3 at line 250, as follows: ' Figure 3 shows vapour pressures above pure, flat, supercooled liquids for n-alkanes in the range C16H34-C32H66, following Chickos and Lipkind (2008), Compernolle et al. (2011), Lemmon and Goodwin (2000), the Epi Suite calculator (US EPA, 2017), and the UmanSysProp tool (Topping et al., 2016).'

P.10, line 237: A-a is mentioned before it is defined in the text. Answer: The text, now at line 261ff, reads: 'An enormous difference in the vapour pressure for C32H66 (from 2.66x10-5 Pa in Epi Suite, to 3.20x10-15 Pa in Nannoolal et al., 2008 with the boiling point of Joback and Reid, 1987, called A-a hereafter) is clearly seen in Figure 3.' ...and at lines 267ff: ' For the purpose of our sensitivity study, three representative datasets are nominated as input, namely: Myrdal-Yalkowsky (1997) with the boiling point of Nannoolal et al. (2004, called B-c in Figure 3 and hereafter); Compernolle et al. (2011, called Co); and A-a.'

P.11, line 270: Please add the abbreviation (Co) for this vapour pressure dataset here. Answer: Done.

P.12, line 291: Here the concept of threshold modal composition is introduced for the first time. It would be good to add a paragraph on the reasoning behind this concept. Answer: We have expanded our description at lines 318ff, as follows: 'For a modal composition of C21H44, increasing $\sigma$ makes almost no difference to the model outcome at 1 s. Below, we call the modal composition that shows insensitivity to $\sigma$ for a given model output time, the threshold modal composition. The threshold modal composition points to the composition compound that is in equilibrium between gas

and particulate phases for the selected timescale. Lower-carbon-number compositions than the threshold modal composition evaporate quicker and therefore have reached equilibrium with their respective gas concentrations on a much shorter timescale. The higher-carbon-number compositions evaporate slowly and are out-of-equilibrium with their respective gas concentrations for the selected timescale.'

P.15, line 358: Although the model results with the vapour pressure dataset Co are explained in detail in section 3.1, it would help the reader to give a short summary of Co results regarding the REPARTEE-like behaviour before A-a and B-c results are discussed. Answer: We have added the following (lines 385-387) 'Diameter change when using Co vapour pressure has been discussed in the previous section. The values of vapour pressure in the Co data are intermediate between the B-c and A-a data. Hence, Dpg,nuc at 100 s using vapour pressure parameterisations A-a and B-c (see Supplementary Information), as expected, shows the same general behaviour as for vapour pressure parameterisation Co, but with a marked change in threshold modal composition.'

P.19, lines 472-477. There seems to be something wrong with the logic of the two sentences. The second sentence contradicts with the statement on p.30, lines 492-493. Answer: We thank the reviewer for spotting this discrepancy. The word greater is replaced with the word lower to comply with the logic of the statements as follows at lines 517ff: 'For components with volatility less than that for the C22H46 surrogate compound used here, all available vapour pressure parameterisations render these compounds volatile over the 100-s timescale. These components will equilibrate with the gas phase on these short timescales. Components with volatility lower than that of the C24H50 surrogate are effectively non-volatile over this timescale for all vapour pressure parameterisations, and so will remain condensed and out-of-equilibrium with the gas phase on these timescales.'

Figure 1: Contours of the GCxGC chromatogram are hardly visible due to the overlay with the coloured polygons. For clarity, it would be better to show the original chro-

matogram beneath the current figure plot. Annotate x-axis and y-axis of the upper bar charts Answer: The coloured polygons in this Figure illustrate that the carbon number distribution does not increase in the same way for different homologous series with increasing retention time (volatility). For example, a C15-alkane has the same volatility as C14 monocyclic alkanes, C13 aldhydes and ketones and C14 monocyclic aromatics. Removing these polygons will not demonstrate this point and the original chromatogram without the polygons will be less informative than is presented in this figure. The polygons provide quick view of the volatility (y-axis) in the homologous series. The contour plot provides the peak intensity and not the mass concentration on the chromatogram. The bar chart on top indicates the total alkane and total mass concentrations per volatility bin, where the x-axis is retention time like what is labelled in Figure 1. The y-axis is percentage mass concentration, which has been added to the Figure.

REVIEWER #2. This reviewer cannot agree with the shrinkage of diesel particles in urban air undergoing evaporative when advected to a cleaner atmosphere, as presented in this study and those reference papers cited. The reviewer also has no solid reason to reject the hypothesis. Supposed that the hypothesis is correct, the modeling results delivered in this study are valuable for research community. Answer: we thank the reviewer for their open-minded reading of our ms.

Two issues might be helpful for potential readers: 1) the temperature change in the initial few second dilution and how this affects particle number size distribution; Answer: Please see the discussion of review #1 above. We do not consider expanding and cooling exhaust plumes, but rather study the neighbourhood-scale transport of the diluted exhaust. We have amended the text at line 138-139 to remind the reader that the simulation is focused on events after dilution and cooling.

2) How does the Kevin effect affect the hypothesized evaporation? Answer: This information is added in section 2.1 (lines 213ff) as follows: 'The Kelvin effect is also considered, which alters the saturation vapour pressure of the compounds as a func-

tion of the particle diameter, the surface tension of the SVOC mixture/solution, and the molecular weight of the participating compounds. The Kelvin effect is pronounced for particles with a diameter less than 20 nm and substantial for particles with diameter less than 10 nm. The Kelvin term accelerates the evaporation for all compounds under consideration in this study and more notably for the high-molecular-weight compounds due to their larger molar volume.'

REVIEWER #3.

Nikolova et al. have used a numerical model to simulate the evaporation of ultrafine particles (UFP) from diesel exhaust under a range of input conditions and have used the model results to develop insight into the organic composition of these UFPs. By informing their model with inputs based on field measurements, they find that certain combinations of model inputs (e.g., <5% of the UFP material can be treated as a non-volatile core) are able to reproduce the observed change in the mode of these UFPs. The motivation for this study is clear and the results are presented in a clear and consistent manner. However, there are several deficiencies in the description of the methods and possibly also in the processes the authors may not have included in their model. Answer: we thank the reviewer for their comments of our ms.

In my view, the authors have undertaken a rather narrow study but need to expand it if they are too truly probe the variable space that can help explain the observations. At this point, I do not recommend publication in ACP until the authors have had a chance to review and respond to my comments. Answer: We believe that our study is of sufficient depth and general interest to warrant publication, and is further enhanced by our answers to the comments below.

Major comments: 1. Organic species distribution: The authors have assumed a Gaussian distribution to model the organic species distribution. Is it possible that the distribution is bimodal? Also, there are plenty of volatility distribution data for diesel exhaust that could be used to inform the type of distribution (e.g., Gaussian) as well as the

spread of the distribution (e.g., standard deviation>X); e.g., Robinson et al. (2007), Grieshop et al. (2009), May et al. (2013). These data could also be used to eliminate certain input choices. Answer: We thank the reviewer for pointing to the references. Recall that our strategy is to scan the vapour pressure and composition distribution and âĂŤ subsequently âĂŤ compare with field and laboratory studies. We agree that those studies show a Gaussian-type distribution centred at volatile SVOC with quite wide standard deviations. Therefore, we have included these references in our discussion as follows (lines 501ff): 'The work of Robinson et al. (2007), Grishop et al. (2009) and May et al. (2013) also point to a Gaussian-type distribution of the exhaust particle composition centred at SVOCs, that has a wide standard deviation.' Our strategy takes a Gaussian approach to the definition of the composition. We cannot, therefore, rule out multi-modal or other distributions that differ strongly from a Gaussian. We have drawn the reader's attention to this limitation of our study at line 182ff: 'Multi-modal compositions, or others differing strongly from Gaussian, are not investigated in the present study, but could be accommodated by a simple extension of the method.'

2. Model details: Several aspects of the model framework are unclear since they are not reported in the main text. Does the model simulate coagulation and nucleation? If yes, how is it dealt with? There is plenty of evidence, particularly with secondary organic aerosol systems, that diffusion limitations in the condensed phase could extend evaporation timescales and the time required for semi-volatile material to reach equilibrium as well as change the dynamics of how an aerosol size distribution evolves over time (e.g., Shiraiwa and Seinfeld (2012), Zaveri et al. (2017)). Is this modeled here? If not, this would be an important process to include. Is the Kelvin effect considered? If yes, how is it modeled? If not, why is not modeled? How does dilution affect the results? Could dilution be modeled to examine the sensitivity in the findings? I have not read the original measurement papers but were there any tracers (e.g., CO) that could be used to account for dilution of the Lagrangian box? Answer: This study does not consider coagulation and the information is now added in lines 208ff : 'Our earlier work (Nikolova et al., 2016) has shown that deposition and coagulation have a minor effect in

the current scenario and so were switched off to allow a more straightforward diagnosis of model behaviour.' Nucleation is not considered because the model simulations are focused on events after dilution and cooling (lines 138ff). Secondary organic aerosol formation and oxidation of SVOC are neglected due to the large timescale in comparison to the timescale in our study. This is now explained in lines 279ff as follows: 'For hydroxyl (OH) radical concentration $\sim$106 molec cm-3, the timescale for atmospheric oxidation of $C16H34$ is about 106 s (Atkinson and Arey, 2003). Therefore oxidation of SVOC is neglected given the timescale in our study (100 s).' The model considers the Kelvin effect. This is explained now in lines 213ff: 'The Kelvin effect is also considered, which alters the saturation vapour pressure of the compounds as a function of the particle diameter, the surface tension of the SVOC mixture/solution, and the molecular weight of the participating compounds. The Kelvin effect is pronounced for particles with a diameter less than 20 nm and substantial for particles with diameter less than 10 nm. The Kelvin term accelerates the evaporation for all compounds under consideration in this study and more notably for the high-molecular-weight compounds due to their larger molar volume.' Dilution has been considered for gas-phase concentration of the participating compounds in the box whereas background gas-phase concentration is kept zero (lines 281-2).

3. Model inputs: Similar to details about the model framework, certain aspects about the model inputs are unclear too. Is the same organic species distribution assumed for all particle sizes? What is the rationale for that? Do the model results change if the organic species distributions are size dependent? Related to the size-resolved composition question, is it also important to consider how the nucleation mode particles measured in the street canyon came to being? For example, if they are formed from the cooling of diesel exhaust as it leaves the tailpipe, could one envision a size dependent composition of the condensing species where the lower volatility species condense on smaller sizes from diffusion limited growth while the semi-volatile species condense on larger sizes from volume limited growth. Is it possible to measure the condensation of the hot/warm tailpipe emissions with this model before it is used to

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model the evaporation? What organic aerosol concentration was assumed to model gas/particle partitioning? Answer: The same organic compound distribution is used for all particle sizes, however, the non-volatile fraction is different between the modes. Aitken mode particles are considered predominantly non-volatile (90% of their mass in non-volatile) and therefore their slow dynamics is of little interest. Nucleation mode particles (diameter less than 30 nm) and their composition, however, are still not fully understood. In this study we found that nucleation mode particles composed predominantly of highly volatile SVOC can explain the shrinkage behaviour seen in the observations of Dal'Osto et al (2011) in Central London. Our model is not capable of simulating exhaust hot tailpipe emissions, nor processes in the first stage of dilution with cleaner air. The organic aerosol mass fraction is based on the Gaussian distribution (please refer to our Supplementary Information) and the total mass per size bin is estimated based on the observed number size distribution in Dal'Osto et al. (2011).Gas-phase concentration is based on the work of Harrad et al. (2003, see the reference list in the ms).

Minor comments: 1. Line 44, 62: No need for citations in the abstract Answer: Because our study links closely to the two previous papers cited in the abstract, and the abstract must be able to stand alone, we respectfully request to retain sufficiently detailed citations, in line with best practice.

2. Line 89: Perhaps define SVOC using the effective saturation concentration definition (C*) of Donahue et al. (2006) too? Answer: This section now reads (lines 89-92): 'Progress has been made in modelling traffic-generated particles (including the ultrafine fraction) using a volatility basis set, defined using the effective saturation concentration (Donahue et al., 2006). Progress in identifying the precise chemical composition of traffic-generated particles has been made by resolving the so-called 'unresolved complex mixture' (largely uncharacterised organics in traditional gas chromatography) via two-dimensional gas chromatography...' Donahue et al., 2006, has been added to the references.

3. Line 107-110: Mention timescales over that 650m too? Answer: We have added the following information on the travel time as follows (lines 118-119): 'The travel time, depending on the wind speed, can vary from about 100 s to 300 s.'

4. Line 117: How are the nucleation and Aiken modes defined? Answer: We have added a description of the modes and the diameter size ranges in section Introduction, lines 75-77, as follows: 'Hereafter, nucleation mode particles are defined as particles with diameter less than 30 nm, Aitken mode particles have a diameter in the range 30 – 100 nm.'

5. Line 155-179: Is it possible that the organic species distribution is bimodal? Answer: See answer to major comment 1, above. We cannot rule out multi-modal compositions where they are not well approximated by a single Gaussian. Our mono-modal organic compounds distribution is in line with the work of Alam et al. (2016) who observed mono-modal dustribution at the tailpipe (please refer to Figure 1 in our ms), and the additional references cited above.

6. Line 283: C20H42? Answer: We do mean C20H42, but this is plotted with the green solid line not the purple. This is now corrected at line 307 as follows:' . For example, at $\sigma$ = 1 and initial mass distribution centred at C20H42 (green solid line with a square marker), . . .'

7. Line 311: I am not sure I understand the comment about longer evaporation timescales for higher carbon number species. My understanding is that the higher carbon number species are already in equilibrium since they have a much lower C* and they would only evaporate if the parcel was continuously diluting – which is not how the model in this work was setup. Can you clarify? Or are you referring to the fact that for two semi-volatile species, the larger species on account of its larger molecular weight/diffusion coefficient would evaporate slower? Answer: We refer to the fact that for two semi-volatile compounds, the largest in molecular weight compound would evaporate slower in comparison with the lightest in molecular weight compound.

C14

We have re-phrased the sentence in our manuscript at line 330ff: 'Furthermore, the timescales are much shorter for those lower than C21H44 carbon-number compositions (e.g. C20H42, C19H40, . . .) and much longer for those higher than C21H44 carbon-number compositions (e.g. C22H46, C23H48,. . .).

8. I would have preferred to use non-volatile instead of involatile in the manuscript, based on the word choice see elsewhere in the literature. Is there a reason for using involatile? Answer: There is no reason for using the word involatile. Literature studies use both words. For consistency with our previous work, all words 'involatile' are replaced with the word 'non-volatile' in our main and supplementary papers.

9. Line 336-343: What are the implications of this section? Answer: This section aims to provide the reader with supportive information on the meaning of threshold modal composition based on our definition and approach.

10. Line 419: The authors mention the use of a size-resolved composition. However, I don't see this information visualized or tabulated anywhere in the manuscript. What am I missing? Answer: All input information is presented in the Supplementary Information. The size-resolved composition can be found in Tables 1S-4S.

11. Line 483: As mentioned above, why wasn't the effect of dilution simulated in this work? Answer: Mixing of cleaner urban background air into the simulated air parcel would lower partial pressures and increase evaporation rates. In our study the gas-phse SVOC data of Harrad et al (2003) is used to represent the urban air while the urban background gas-phase concentration is set to zero therefore observing a maximum decrease in particle diameter.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-1018/acp-2017-1018-AC1-supplement.zip

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