Atmospheric
Chemistry
and Physics
Discussions

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.

Anonymous Referee #3

Received and published: 28 April 2018

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. 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.

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.

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?

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 model the evaporation? What organic aerosol concentration was assumed to model gas/particle partitioning?

Minor comments:

1. Line 44, 62: No need for citations in the abstract

2. Line 89: Perhaps define SVOC using the effective saturation concentration definition (C*) of Donahue et al. (2006) too?

3. Line 107-110: Mention timescales over that 650m too?

4. Line 117: How are the nucleation and Aiken modes defined?

5. Line 155-179: Is it possible that the organic species distribution is bimodal?

6. Line 283: C20H42?

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?

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?

9. Line 336-343: What are the implications of this section?

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?

11. Line 483: As mentioned above, why wasn't the effect of dilution simulated in this work?

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