Interactive comment on “Modelling and measurements of urban aerosol processes on the neighborhood scale in Rotterdam, Oslo and Helsinki” by M. Karl et al.

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We thank Referee #1 for positive evaluation of the manuscript. Following the Reviewer’s remarks, significant changes will be made in the revised manuscript in order to improve the description of the PN measurements, as well as the discussion of uncertainties of the simplified PN parameterization and the consideration of nanoparticle loss by evaporation.
1. Line 15 of the Abstract. "It was not necessary to model the nucleation of the gas-phase vapors..." This sentence created immediate concern (suggestion nucleation had been neglected) until I read further into the manuscript to understand fully. I suggest adding a short sentence after this to say why, i.e. post tail-pipe emissions were considered.

Response:

The sentence will be changed to: "By choosing concentrations and particle size distributions at roadside as starting point of the computations, nucleation of gas-phase vapors from the exhaust has been regarded as post tail-pipe emission, avoiding the need to include nucleation in the process analysis."

2. Line 22 of pg 35168. "The traffic volume at Bentinckplein, which is a street canyon..." The measurement PN in Street canyons is affected by the circulation of air within the canyon whether it passes across the traffic before passing over the sampler or whether it passes over the sampler first before the traffic. This is dependent on the wind direction across the canyon. Was this taken into account?

Response:

When modelling the hourly contribution from road traffic emissions to the sampler location in Rotterdam at the Bentinck street location, the hourly average wind direction was indeed taken into account.

The following sentence will be added on p. 35168: "Hourly averaged wind direction was used to select campaign data that was directly influenced by the traffic emissions in the street."

3. Line 21 of pg 35171 "The mean traffic-related size..." Three distinct modes are described with mean diameters 17, 85 and 250nm. Are these peak fitted modes and if so please specify how you fitted them.
Response:

Three-modal fit has been done with the MAFOR model; the procedure is explained in section 2.2 of the manuscript. This information will be added.

4. Lines 1-10 of pg 35184. In the conclusions the inaccuracies of the simplified treatment of the coagulation process was discussed, which did not account for the coagulation between size categories. How does the accuracy improve as the number of size categories is increased, eg X2 and X4?

Response:

We note here that the discussion of the accuracy of the derived simplified parameterizations of dry deposition and coagulation involving three size categories of PNC only elaborates on uncertainties with respect to the solution obtained from the fully size-resolved MAFOR model with 120 size sections. Uncertainties due to the neglect of fractal geometry and van der Waals forces as well as uncertainties due to inaccurate measurement of dry deposition velocities, as pointed out by Referee #2, are not included in the error of the simplified parameterization. The accuracy of the developed parameterization improves when the number of size categories is increased since the error due to neglecting intra-modal coagulation is reduced. For a parameterization with six categories of PNC the deviation to the reference solution with 120 size sections is only up to 5 %. Further increasing the number of size categories is expected to give even better accuracy however is not considered to be practical for the implementation in urban dispersion models.

The following sentence will be added to section 3.7 "The recommended simplified parameterizations of aerosol processes" in the revised manuscript: "Increasing the number of PNC size categories is expected to reduce the error due to neglecting coagulation between size categories. A parameterization with six PNC categories resulted in a deviation to the reference solution by only up to 5 % (Table S3). In addition the parameterization is uncertain due to assumptions about particle shape, neglecting van der
Waals forces as well as inaccurate measured dry deposition velocities. It is however not affected by the specific treatment of dilution in the idealized scenarios because the simplified PNC parameterization was derived with only one aerosol process activated.

A table with obtained with required data for implementation of the 6-category parameterization will be included in the Supplement (new Table S3).

5. Table 3 pg35193 The average PN concentration for Helsinki LIPIKA Case 1 (186100 cm$^{-3}$) is much higher than the other sites, what is the explanation for this?

Response:

We thank Referee 1 for finding this mistake. The high value of 186 100 particle cm$^{-3}$ was measured with the mobile laboratory "Sniffer", while driving on Itäväylä, in 9 m distance from the center line of the two-direction highway, and not as erroneously written in Table 3, at 65 m distance from roadside. The measured value in 9 m distance from the roadside was 129 600 particles cm$^{-3}$. This is the correct value, which has also been used in the calculations. Table 3 and Figure S1 will be corrected accordingly.

6. Figure 4A. The size development of the size spectra for Oslo Winter UFP - Oslo shows the nucleation mode being diluted. It would be useful to have had a third intermediate size distribution collected half way between the roadside and background to fit the model through. Is there a possibility that there could be nanoparticle loss by evaporation, rather than coagulation processes? (ref M. Dall'Osto et al Atmos. Chem. Phys., 11, 6623-6637, 2011).

Response:

We agree that intermediate sampling positions in 100-1000 m distance between the roadside and the background site would have been of great value to determine possible evaporation of exhaust particles.

We give a detailed evaluation on the possibility of loss by evaporation in our response...
to the Comment by Prof. Harrison and co-workers.

The title of section 3.4 will be changed from "Effect of condensation of n-alkanes" to "Effect of condensation and evaporation of organic compounds".

The following text on findings from Dall'Osto et al. (2011) and Harrison et al. (2016) will be added in section 3.4:

"Model simulations of the idealized scenario suggest that evaporation could be an important process, altering the particle size distribution in urban micro-environments, if the semi-volatile vapor and also the nanoparticles forming post-emission were assumed to have the same or higher volatility as the n-alkane C22. Dall'Osto et al. (2011) analyzed observations of particle size distributions from London and reported a reduction in the size of nucleation mode particles during advection from a major highway into the cleaner environment of a park, indicating evaporative loss of semi-volatile constituents during travel times of around 5 min. Harrison et al. (2016), for the same location, found most rapid evaporation to occur at higher wind speeds, associated with shorter travel times, but cleaner air."

References:


Interactive comment on Atmos. Chem. Phys. Discuss., 15, 35157, 2015.