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Discussions

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Interactive comment on "Downwind evolution of the volatility and mixing state of near-road aerosols near a US interstate highway" *by* Provat Kumar Saha et al.

Anonymous Referee #2

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The authors present a dataset of aerosol evaporation in thermodenuders and ACSM and black carbon measurements measured at several distances from a major highway. The dataset and the associated analysis of the volatility and the mixing state of the aerosol at different distances downwind of the road is very valuable to atmospheric aerosol and traffic emission researchers. The measurements are to my knowledge the first time the volatility and mixing state have been measured at several points along the roadside, and as such the dataset is novel and interesting, and the subject area is clearly in line with ACP. The manuscript is well written and the data analysis is comprehensive; there are some issues that I suggest are addressed in a revision, after which I recommend publication.

1. As the traffic-originated aerosol is transported away from the source, it dilutes with background aerosol with a rate that depends on the wind velocity and the atmospheric stability. Therefore, the same measurement point (measured by distance) can see aerosol with different age since emission, and different dilution factors. This can be be overcome by measurements of some relatively inert gas at the roadside and downwind, or more crudely just estimating the aerosol 'age' from the wind velocity, distance (and direction).

Neither of these have been performed in the manuscript, and therefore the measurement at each distance may include differently aged particles, with a varying fraction of background aerosol mixed in the traffic-originated aerosol. This impacts the generalization of the results (which may easily happen, given the scarcity of this type of data in the literature), and I think that this should be made clear to readers. This could be done e.g. by providing estimates, and ranges of variation, of the age of particles as an additional variable. Also, this would give more confidence in whether the observed seasonal differences are due to actual differences in volatility or maybe just different mixing situations.

2. Comparing the distributions in Fig S3, it seems that an important factor in the change of volatility is the disappearance of a large fraction of particles smaller than ca 40-50 nm between the measurement points at 10 m and 150 m, and this seems to contibute to the large change in volatility of <100 nm particles (the change seems much less for the 100-400 nm particles). From this one could assume that these small particles are mostly consisting of SV particles. On the other hand, these smallest particles are most affected by e.g. coagulation, and their mass could thus be transferred to the larger size range during transport. I think that the large change with transport in the <50 nm particle size range should be investigated, and maybe some overview of the literature on whether this is a typical occurrence could be added to the paper.

Some other minor comments

page 8, line 15: 'Other effects (if any)...' This sentence is mostly confusing; either the other effects are clearly mentioned here, or this sentence could be removed.

page 9, line 1: I'm not following what is meant by 'downwind trend is dictated by BC'. What is the BC-related process that determines the VFR? Also, it is not clear what the quantity VFR-OA is representing here. How is this related to the difference in BC and denuded SMPS volume in Fig. 2d? Also, is fig 2d an example or representative of a longer period? Please clarify.

Page 9, line 15, and Fig 3 and 5. The sentence 'The evolution of the size distribution of a monodisperse particle upon heating' is confusing. Also, how is the color scale chosen in Figs. 3 and 5? As the main information of the figure is the relative contributions of the different modes, could it not be useful to normalize the volatility spectra? Now the 25 nm spectra are very vaguely readable.

page 11, line 6: '...the mixing state of traffic-emitted particles is not substantially altered within a few hundred meters'. This is basically correct in the context that particles that are externally mixed at the start stay so; however, in a hypothetical case that internally mixed particles dominate the emission at the start, the mixing with background air would soon cause an external mixture. The sentence should be made less general, e.g. '...in this case, the mixing state...'

page 13, line 26, and Fig. 8. I have some difficulties understanding how Fig. 8b was arrived at, and what should be compared in the figure. Which bars at at 10m and which ones at 220? The statement 'we assume that the volatility distribution (...) at 220 is a representative (...) for background particles' seems contradicting to fig 8a, where ca 33% of the total aerosol seems to still be traffic-originated. I think that it would be helpful to understanding if a more detailed (step-by-step) explanation for how each of the three mass concentrations were obtained could be given, maybe even in equation form in the supplementary. For example:

* $M(\text{near-road})_i = (\text{SMPS total mass} - BC - \text{ACSM(inorganics)}) \times (\text{May et al})_i$

C3

* $M(\text{background})_i = (\text{bg OA mass}) \times (\text{TD vol. distribution at 220m})_i$

* $M(\text{traffic})_i = (NR - BG \text{ oa mass}) \times (\text{TD vol. distribution at 10 m})_i$

Several assumed densities could be found in the article for different data analysis approaches (at least 1.1, 1.5 and 1.8 g/cm^3 are described). Could this be made more consistent, or are there specific reasons for using these values that could be stated?