

Responses to Reviewers' Comments on Manuscript ACPD-2018-844

(Estimating the influence of transport to aerosol size distributions during new particle formation events)

We thank the reviewers for their comments to improve this manuscript. We have addressed the comments in the following paragraphs and made corresponding changes in the revised manuscript. Comments are shown as *blue italic text* followed by our responses. Changes are highlighted in the revised manuscript and shown as underlined text in the responses.

Reviewer #1:

This is a very well-written manuscript dealing with an important topic: the effect of transport on interpreting new particle formation (NPF) events using particle number size distribution (PNSD) measurements. Although it is well known that PNSDs are affected by inhomogeneities in measured air masses, no proper tools to take this into account in analyzing NPF event have been proposed so far. This manuscript addresses this topic. I have couple of issues that the authors could discuss a bit more in the paper and a few other relatively minor comments. After the revisions, the paper should definitely accepted for publication.

Main issues

The authors do not comment anything about the width of the considered size range $[d_i, d_j]$. It is clear that there are both benefits and drawbacks of using either a narrower or broader size range. For example, a broad size range would worsen some of the assumptions stated on page 6, such as the influence of primary emissions and constancy of the particle growth rate (GR). A very narrow size range would probably cause more noise into some of the terms that influence the calculation of TR from equation 10. Please discuss shortly this issue in the paper.

Response: Thanks for the suggestion. We addressed the selection of the analyzed size range and the corresponding reason in the revised manuscript and supplementary materials. We also move Eq. 11 and the relevant paragraph on converting the transport term into the cumulative transport term to Section 2, because integrate the transport term with respect to time can also reduce the influence of random measurement uncertainty in addition to using a broad size range. The relevant paragraph is revised as:

“The estimated transport term usually needs to be properly smoothed to reduce the impact of the uncertainties in the estimated $dN_{[i,j]}/dt$, $CoagSnk_{[i,j]}$, and the growth term. Using a wide size range of the concerned size bin $([d_i, d_j])$ can help to reduce measurement uncertainties. Alternatively, the transport term can be integrated with respect to time. The temporal evolution of particle number concentration ($dN_{[i,j]}/dt$) and the other terms in Eq. 10 can be converted into the temporal evolution...In addition to reducing the impact of the fluctuations in the observed aerosol size distributions due to measurement uncertainties, these transformations facilitate the comparison

among different size bins. In this study, we use a single measured size bin to analyze the contribution of transport to minimize the systematic error caused by the assumption of a size-independent GR. The representativeness of an analyzed size bin are tested by comparing to its adjacent size bins, as the example shown in Fig. S1.

As demonstrated by the equations derived in this paper, knowing the particle growth rate (GR) is needed to estimate the transport effect on NPF (the same concerns also calculating other relevant quantities related to NPF like the particle formation rate). The authors need to assume a constant GR to apply equation 10. I have a few comments related to this. First, the authors state on page 6 that constant GR is a good assumption over the size range 10 to 50 nm. This is only true if particles are growing by condensation of essentially non-volatile vapors. A number of studies have reported a strongly size-dependent GR in the sub-20 nm size range, and usually explained this feature either by more and more volatile organic vapors being able to condense onto particles as they get bigger. Furthermore, any contribution to GR from heterogeneous processes in growing particle would probably make GR not constant with particle size. This may be important, as e.g. Paasonen et al (2018, Atmos. Chem. Phys. 18, p. 12085) showed that, in long-term data from one measurement site, the average GR increased by about a factor 3 from the particle diameter of 10 nm to the diameter of 100 nm. Second, in cases where the transport effects are most important, it may either be very difficult to determine GR from measured PNSD data or, in case GR can be determined, it might not reflect the real GR of the measured particle population. The authors should bring up these issues and also comment shortly whether, and in which cases, they would cause problems in determining TR from equation 10.

Response: In the Theory section, we added:

“The accuracy of the estimated transport term is affected by the uncertainty of the estimated growth rate. Equation 10... Even in the recommended size range, 10-50 nm, particle growth rate may sometimes be size dependent due to the uptake of semi-volatile vapors (Paasonen et al., 2018). Accordingly, we recommend to use a narrow size range for estimating the transport term to minimize the potential systematic error caused by size-dependent growth rate. The change in particle diameter due to transport may sometimes contribute significantly to the estimated growth rate. For instance, a diameter shift in particle diameter due to a sudden shift in wind direction may be mistaken as condensational growth if using a time-resolved growth rate. It is usually difficult to decouple the particle diameter shift due to transport and condensational growth rate because the contribution of transport is usually assumed negligible when estimating the growth rate. Accordingly, we recommend to determine growth rate via fitting particle peak diameter over a time range only when the wind speed and direction are relatively stable. Although this fitting method does not decouple the influence of transport, it may help to reduce uncertainties in the estimated growth rate. In addition, if there is a significant error in the method to estimate the growth rate (Li and McMurry, 2018), the error will also propagate into the estimated transport term.”

In section 4.4 (Remarks on the feasibility of the balance method), we added “The errors in the estimated CoagSnk and the growth term also contribute to the uncertainties in the estimated transport term”.

Minor issues

In principle, all figures should be cited in a numerical order in the text. This is not the C2 case for Fig. 3a on line 14, page 6. However, in this case referring to figure 3a there is understandable as it is given as an example and then treated in more detail later in the text. To make this clear, it would replace "(see Fig. 3a)" on this line with "(see Fig. 3a in section 4.1)".

Response: We revised “(see Fig. 3a)” as “(see Fig. 3a in section 4.1)”.

There is a large body of literature on liquid-phase reactions in aerosol particles after the study by McMurry and Wilson (1983) cited on line 5, page 7. I would recommend adding one or two more recent papers into here.

Response: We added Moch et al. (2018) and Song et al. (2018). This sentence was revised as “...liquid phase reactions are possibly an important mechanism for particle growth (e.g., McMurry and Wilson, 1983; Moch et al., 2018; Song et al., 2018), indicating...”.

Grammatical issues:

page 2 line 24: . . . , no dramatic increase . . . was observed, . . .

page 2, line 4: . . . contribution . . . to . . .

page 8, line 24: . . . at around 10:30 . . .

page 10, line 30: . . . were formed. . .

Response: Thanks, corrected.

References

Moch, J. M., Dovrou, E., Mickley, L. J., Keutsch, F. N., Cheng, Y., Jacob, D. J., Jiang, J., Li, M., Munger, J. W., Qiao, X., and Zhang, Q.: Contribution of Hydroxymethane Sulfonate to Ambient Particulate Matter: A Potential Explanation for High Particulate Sulfur During Severe Winter Haze in Beijing, Geophysical Research Letters, 10.1029/2018gl079309, 2018.

Song, S., Gao, M., Xu, W., Sun, Y., Worsnop, D. R., Jayne, J. T., Zhang, Y., Zhu, L., Li, M., Zhou, Z., Cheng, C., Lv, Y., Wang, Y., Peng, W., Xu, X., Lin, N., Wang, Y., Wang, S., Munger, J. W., Jacob, D., and McElroy, M. B.: Possible heterogeneous hydroxymethanesulfonate (HMS) chemistry in northern China winter haze and implications for rapid sulfate formation, Atmospheric Chemistry and Physics Discussions, 10.5194/acp-2018-1015, 2018.