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 <u>underlined text</u> in the responses.

Reviewer #2:

General comments:

This paper deals with an important topic for NPF measurement interpretation: estimating the effect of transport on the observed NPF events. The authors clearly explained how the population balance method can be used to evaluate whether the transport term is significant. Although some comments may be added to strengthen some points made in the paper, the paper is well written and the authors did not overstate their conclusions. I recommend this paper be accepted for publication after the authors address the two comments listed below as well as the issues brought up by the first referee.

Specific comments:

1. My major concern with the current method is the neglect of transport on apparent particle growth: 'the contribution of transport on apparent diameter is neglected in this study' (page 6, line 12). Although it is possible the effect of transport on apparent particle growth is indeed negligible, the opposite may also be true: the observed growth is due to particle growth elsewhere, and these grown particles are transported to the observation site. As pointed out by the authors, for the south-east Tibet observation, the correlation between local wind field and estimated transport terms suggest the particles may have been formed elsewhere (page 10, line 19). It is my understanding that the application of equation 10 requires the growth term to be decoupled from the transport term; while inferring growth from the peak diameter shift does not guarantee this decoupling. I suggest the authors add more comments on this.

<u>Response:</u> We discussed the uncertainty in the Theory section since the current method (Eq. 10) cannot decouple the contribution of transport to the observed peak diameter shift:

"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 a sudden shift in wind direction may be mistaken as condensational growth if using a time-resolved growth rate in Eq. 10. 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 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."

For the south-east Tibet observation, we added "<u>The relatively constant cumulative transport terms</u> of 10 nm and 20 nm particles between 12:00 and 14:00 indicate a relatively small contribution of transport to the observed shift in particle diameter and hence the estimated growth rate." The relatively constant cumulative transport term illustrated in Eq. 11 indicates an averagely near zero transport term in Eq. 10. Thus, transport may not be the reason of the observed particle diameter shift. For other specific events, the influence of transport on apparent particle growth may be significant and we think the newly added paragraph in the Theory section explains this problem.

For the urban Beijing observations, the particle number concentration are quite high. In this case, it is possible that cogulation contributes to the apparent particle growth in this. For this data set, it seems that the authors didn't apply correction techniques, e.g. the method given by Stoltzenburg et al.(2005), to account for the coagulation effect on particle growth. After accounting for coagulation, to what extent will the growth term shift?

<u>Response:</u> We corrected the coagulation contribution to the apparent growth rate, updated Fig. 9, and added "<u>The contribution of coagulation to the estimated grow rate was corrected (Stolzenburg et al., 2005)</u>" in the main text and the caption of Fig, 9. The growth term decreases ~15% after correction. The cumulative transport term is still negligible compared to the cumulative *CoagSnk* and cumulative growth term after correction, as shown in Fig. R1.



Fig. R1: The time series of dN/dd_p , the cumulative transport term, the cumulative CoagSnk and the cumulative

growth term of 30 nm particles before and after the correction of the coagulation contribution to particle growth.

We understand that the accuracy of the estimated growth rate is important to the estimated transport terms. Although coagulation contributes a minor proportion to particle growth during this event, it may be important during other events. Hence, we stated that "in relatively polluted atmospheric environment such as urban Beijing, however, the estimated the contribution of transport is sensitive to the uncertainties in CoagSnk and the growth term" in the original manuscript and listed "particle formation due to coagulation ($CoagSrc_{[i,j]}$) is negligible compared to particle loss due to coagulation scavenging ($CoagSnk_{[i,j]}$) and condensational growth (characterized using the growth term)" as an approximation of the current method.

Technical corrections:

page 1 line 28: ... in a relatively clean atmospheric environment... page 9 line 23: ...at different particle diameters are correlated with... page 11 line 30: ... in a relatively polluted environment...

Response: Thanks, corrected.