

Author's Response

By M. Kulmala (Referee #1)

1) Page 15152, line 11 ... few nanometer ... This is not correct. Size of atmospheric particles starts from ca 1.5 nm (see Kulmala et al. Science, 2013)

Response:

As suggested, we have now changed the size range from few nanometer to 1.5 nm and the amended text reads as: “*Size of atmospheric particles, ranging from 1.5 nm to hundreds of micrometers, is a key factor for evaluating environmental effects of particles (Kulmala et al., 2013; Buseck and Adachi, 2008; Kumar et al., 2014)*”.

2) Page 15153. It is important to add references to recent nucleation studies made by Hermann et al. (2014, ACP) as well as other studies performed at SORPES station in Nanjing.

Response:

We thank the referee for pointing out this station and the reference to us. We have now discussed this reference and the amended text reads as: “*Though the first study on NPF events during polluted episodes was conducted in the megacity of Beijing (Wehner et al., 2004; Wu et al., 2007), the occurrence of NPF events is only reported at a few sites in China up to now (Du et al., 2012; Liu et al., 2008; Wang et al., 2013c; Wehner et al., 2004; Wiedensohler et al., 2009; Herrmann et al., 2014)*”.

3) Page 15159. Authors should give references for equations 3 and 4. E.g Kulmala et al. 2012 (Nature Protocols)

Response:

As suggested, we have now cited the above-mentioned reference as an original source of these equations. The text now reads as: “*The growth rate (GR) of newly formed particles and condensational sink (CS) are calculated for NPF events. The CS determines the how quickly of the gaseous molecules can condense onto the pre-existing aerosols and can be calculated by using Equation (3) (Kulmala et al., 2001, 2012)*” and “*GR is calculated using the Equation (4) (Kulmala et al., 2012)*”.

4) The instruments start to measure at 15 nm. Since it takes up to several hours to reach that size from nucleation (ca 1.5-2 nm), it would be good to discuss where and when nucleation have been occurred.

Response:

We thank the referee for his clarification on the nucleation process. As suggested, we have added the following text to address this point: “*As the lower cut-off diameter of our measurements was 15 nm, which is much larger than the size of nucleation clusters (1.5-2 nm) (Kulmala et al., 2013), the NPF events might occur 1 to 2 hours before we*

observed them”.

5) Page 15165: why there is not NPF if air mass is coming from ocean?

Response:

At coastal sites, no NPF events were observed to take place when the air mass was coming from the ocean side with clean air to the measurement station (e.g., WL_c, BG_c). This is because the cleaner air mass was not carrying enough NPF precursors such as H₂SO₄ or low-volatile VOCs to favor the nucleation events. We have now modified the text in lines 404-411 to make the above point clearer and the amended text reads as: “*At coastal sites, however, no NPF events were noted when the air mass was coming from the ocean side with clean air, as this cleaner air mass was not carrying enough NPF precursors such as H₂SO₄ or low-volatile VOCs to favor the nucleation events. This is substantiated by the fact that SO₂ concentrations in the air coming from ocean side were much lower than those coming from the continent (e.g., at BG_c site, average SO₂ concentration in the air mass from ocean and continent were 1.4 ppbv and 2.8 ppbv, respectively) and supported by other studies at coastal site (Yu et al., 2014)*”.

6) Page 15166 line 9. GR one time higher? I do not understand this.

Response:

We have now made this statement clearer which now reads as: “*Average GRs at urban and regional sites were about twice those at coastal sites and cruise measurement, indicating that the higher concentrations of gaseous precursors in the polluted areas not only favor the formation of particles, but also accelerate the growth rate as long as the nucleation particles are formed*”.

7) Page 15168: How many days or hours it takes from NPF to CCN? It is crucial to look whether CCN concentrations are increasing in the second or even in the third day.

Response:

The time it takes from NPF to CCN depends on the growth rate (GR) of nucleation particles. In this study, we find that the GRs of nucleation particles at urban and regional sites were 3.2 nm/h to 21 nm/h, which means that it takes roughly about 2.5 to 16 hours for the nucleation particles to perform as CCN (assuming the critical diameter is 50 nm). On the other hand, the GRs of nucleation particles at coastal sites and cruise measurement were 1.6 nm/h to 7.5 nm/h, suggesting that it will take much longer time (roughly one day) for the nucleation particles to reach CCN size range (at least 50 nm). The referee have rightly pointed out that it is crucial to evaluate the contribution of NPF events to CCN production in the second or the third day. In our study, we use the mode fit method to estimate the contribution of NPF to CCN. This approach requires very clear NPF peaks in the size distributions in order to achieve accurate fitness results. As

a result, we cannot calculate the contribution of NPF events to CCN production in the second or the third day. Nevertheless, to make this much clearer, we have modified the corresponding discussion. The text now reads as *“Third, as discussed above, the approach requires very clear NPF peaks in the size distributions in order to achieve accurate fitness results. So the focusing time period is constrained between 14:00 and 17:00 h, when the NPF mode is clear and evening rush hour has not come. The contribution in the following hours or the following days cannot be precisely calculated though this approach. As it is crucial to evaluate the contribution of NPF events to CCN production in the following days, more studies on this field are needed in the future.”*

8) It would be good to speculate what are formation rates at 3 nm. It is possible to calculate formation rates at 15 nm, although they probable have nothing to do with atmospheric nucleation.

Response:

The referee have rightly pointed out that the formation rate is an important parameter to describe the characteristic of nucleation events, which is even more important to study the mechanism of new particle formation (Kulmala et al., 2013). However, as our measurement of particles starts from 15 nm, the calculated formation rates cannot precisely represent the real nucleation rate at 3 nm or 1.5 nm. This is the reason why we have not discussed them in our study.

By Anonymous Referee #2

This manuscript summarizes the results of particle number size distributions from several field studies conducted in China, and analyses the major implications obtained from these measurements with a special focus on new particle formation and growth and subsequent cloud condensation nuclei production. The paper is definitely of interest for the scientific community. The paper appears to be scientifically sound, with no clear errors in it. I have a few minor and mainly technical issues that should be considered before publication.

1) The authors should be clear and consistent in the way they represent the averages, the ranges of averages and overall variability of the different quantities in the text. For example, the ranges of CS reported at the end page 15165 seem to represent overall ranges of these quantities in different types of stations (see Table 3), whereas the ranges of GR at the beginning of next page represents the corresponding ranges of site-averaged values. Furthermore, the authors have not defined what they exactly mean by these ranges in the tables (minimum to maximum values or some percentage range?).

Response:

As suggested, we have now changed the statement of ranges of GR in the way of overall ranges of the quantities in different types of sites. The text now reads as: *“The GRs of*

newly formed particles (calculated from 15 nm to 30 nm) ranged from 4.2 to 18.1 nm h⁻¹ at urban sites, 3.2 to 21 nm h⁻¹ at regional sites, and 1.6 to 7.5 nm h⁻¹ at both coastal sites and cruise measurement (Table 3)”.

For Table 3, the values outside the bracket represent the average CS or GR while those inside represents the maximum and minimum CS or GR. We have now modified the caption of Table 3 to make this clearer.

2) While the text was quite well written in general, there were some grammatical problems that should be corrected: The use of tense should be carefully checked out throughout the paper. Past tense should preferably used when representing the results or what was done. This seems to be correct in most places of the text, but in some paragraphs the authors use the present tense instead. Sometimes this causes confusion. For example, past tense on lines 11-12 in page 15161 indicates that what is said here is the results of this particular study. However, a reference is added there which indicates that is rather a more general result concerning biomass burning aerosols. Which one do the authors mean?

Response:

We meant that aerosols from biomass burning are generally larger in size and may influence the diameter of measured particles at regional sites. We have now revised the sentence in present tense.

Also, we have now got our manuscript proof-read by a native English speaker to remove any grammatical infelicities.

3) Articles are missing from several places of the text. Please check out and add.

Response:

We thank the referee for pointing it out. We have checked the manuscript to remove grammatical errors.

4) Line 16 on page 15165: procurers?

Response:

Thank you for pointing this typing error. We have now changed the word “procurers” to “precursors”.

5) Line 9 on page 15166: “one time higher than...” sounds strange. Do the authors mean “about twice those in...”?

Response:

Revised as suggested. Thank you for pointing this out. The text now reads as “Average GRs at urban and regional sites were about twice those at coastal sites and cruise measurement, indicating that the higher concentrations of gaseous precursors

in the polluted areas not only favor the formation of particles, but also accelerate the growth rate as long as the nucleation particles are formed”.

6) The format of giving variable ranges is incorrect in some places of the text. Correct way are to state ... ranged from M to N ... were in the range M-N, or ...were between M and N.

Response:

We thank the referee for pointing it out. We have checked the manuscript to revise grammatical errors in lines 260, 413 and 422 as suggested.

References:

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