

p12496, 118: The hygroscopic growth of particles has been shown to have a significant effect on sink calculations (e.g. Horrak et al., 2008), and should thus be taken into account in the CS calculation (using “wet” diameters instead of “dry” ones) since particles are dried before entering the DMPS.

**Response:** *Thanks for the comment. We will present both CS (with/without the consideration of hygroscopic growth) in Table 1 in revised manuscript. Since there is no observation about hygroscopic growth of particles at SORPES station until now, we will use hygroscopic growth data in another suburban site in north of downtown Nanjing (Wu et al., 2014) following the method of Laakso et al. (2004).*

p12497, 119-21: If particle concentrations from the urban site near Beijing (32 700 cm<sup>-3</sup>) are considered comparable to those measured at the SORPES station (19200 ± 9200 cm<sup>-3</sup>), concentrations from the rural site (11500 cm<sup>-3</sup>) should also be considered as similar, and not lower.

**Response:** *Thanks. We will correct this point in the revised manuscript.*

p12497, 125: I would say “which can be several times higher” instead of “being several times higher”, since based on Table 1 and Fig. 2, concentrations lower than 2900 cm<sup>-3</sup> are reported for the accumulation mode at the SORPES station, especially during summer (5300 ± 4200 cm<sup>-3</sup>).

**Response:** *Thanks. We will make a change according to the suggestions.*

p12498, 11: Which concentration is correct: 5700 cm<sup>-3</sup> from the text or 5800 cm<sup>-3</sup> from Table 1?

**Response:** *5700 cm<sup>-3</sup> was the number concentration in the size range of 100-500 nm. In Table 1, the 5800 cm<sup>-3</sup> was the NC from 100 nm to 800 nm.*

p12498, 13: The definition of the seasons can slightly vary from one study to another, so I think it would be great to clearly state which month belongs to which season to avoid any confusion.

***Response:*** Thanks. We will make a change according to the suggestions.

p12498, l6: Instead of talking about “elevated concentrations in early winter and spring”, I would rather mention peaks in December-January and April, since, concentrations from the other spring months, ie March and May, are not elevated compared to the concentrations observed between July and November.

***Response:*** Thanks. We will make a change according to the suggestions.

p12498, l7: Based on Fig. 2, it is true that from November to June, the month-to-month variability of the Aitken mode concentration is lower than the variability observed for particles in the nucleation mode. However, I would say that the variabilities are comparable during the rest of the year.

***Response:*** Thanks. We will make a change according to the suggestions.

p12498, l24-25: Extra sources, such as domestic heating, cannot give additional explanation to the higher particle loadings in the accumulation mode in winter?

***Response:*** In Nanjing there is no additional domestic heating in winter, which only existed in north China. But of course, it could somehow influence Nanjing through long-range transport. In revised manuscript, we will add some discussions to address this point.

p12498, l1-2: Regarding the sentence: “Radiation connected with NPF events and local directemissions from vehicles influenced the NC of nucleation and Aitken mode particles”. It is quite easy to understand that the seasonal variations of radiation/NPF frequency can, at least partly, explain the variations observed on Fig. 2 b and c. However I do not understand how vehicle emissions can explain such variations. Do these emissions show a significant seasonal pattern? And if they do, can you give an explanation?

**Response:** *The vehicle emission itself didn't have very strong seasonal variation in Nanjing.*

*However, the more stable and lower mixing layer in winter might cause the higher concentrations of nucleation and Aitken mode particles even with the same emission. We will add some discussions in the revised manuscript.*

p12499, 19-13: The sentence is quite long, and the brackets do not ease the reading. Maybe two shorter sentences could help!

**Response:** *Thanks. We will make a change according to the suggestions.*

p12499, 125-27: Regarding the statement: "In winter, direct emissions from vehicles might play a key role in the diurnal cycle of particle number size distributions". Again, I do not understand why emissions from vehicles are believed to show seasonal variations, being stronger in winter.

Moreover, based on Fig. 4, peak concentrations related to the rush hours are not only seen in winter, at least for 100 nm particles. Thus, I would rather say that in winter, when other sources such as regional NPF are weaker, the contribution of vehicle emissions to the total particle concentration is probably higher compared to other seasons, and even enhanced by lower boundary layer heights, as suggested on p12500, 16.

And, again, what about other anthropogenic sources in winter, such as domestic heating?

**Response:** *Thanks. We will make a change according to the suggestions.*

p12500, 12: The fragment "typical seasons" sounds strange.

**Response:** *Thanks. We will make a change.*

p12500, 118: Why did you decide to calculate backward trajectories over two days, and not over a longer time period? This choice is adapted to the study of freshly nucleated particles, which have a turnover time evaluated to be between 1.6 and 1.7 days. However, performing back trajectories over a longer period would be more appropriate if you want to include larger particles into the

discussion (turnover time of 2.4 days for 200 nm particles (Tunved et al., 2005)).

**Response:** *Thanks for the suggestions. 3-day backward trajectories will be presented in the revised manuscript.*

p12501, 11-3: Can you justify the fact that clusters C2 and C3, and clusters C4 and C5, can be considered as one air mass type, ie continental and YRD, respectively? Based on Fig. 6, C3 air masses travel over a large area of BVOCs, which is not the case for C2; C5 air masses seem to be more local compared to C4 air masses, which probably have an additional marine signature. It is even harder for the reader to appreciate if the classification continental/YRD is appropriate since Fig.7 only shows average (mean or median? should be precised) variations, and no variability.

**Response:** *Thanks for the suggestions. In the revised manuscript, we will modify Figure 7 by showing the size distributions and diurnal variations of number concentration of all 5 clusters. C2 and C3, C4 and C5 will be discussed separately. C3 air masses experienced highest level of nucleation mode particles because the air masses appeared mostly in summer and transported over BVOCs abundant regions. As the C5 air masses were more local compared to C4 air masses and C4 air masses went through large area of marine, C4 air masses had lower accumulation mode particles while higher Aitken mode particles than C5 air masses.*

p12501, 14-11: First, I would remove the sentence “With low...daytime”, since it is a general statement that does not highlight the analysis related to the influence of air masses. Moreover, a full analysis of the parameters influencing NPF is proposed in section 3.2.2.

Regarding the statement “because such air masses are always associated with sunny days and low humidity”, I would balance this statement and say “most probably because”, first because the conditions that favour NPF are discussed in detail in section 3.2.2, and second because some BVOCs, which are mostly seen in C3 cluster, may give additional explanation to the high levels of nucleation particle in continental air masses.

Finally, I suggest to show the position of Nanjing on the maps (Fig. 6), so it would be easier to follow the explanation from I9 to I11.

**Response:** *Thanks. This paragraph will be re-written in the revised manuscript and the location of Nanjing will be added in Figure 6b.*

p12501, 118: Why were undefined and non-event days considered all together? I think undefined days should be considered separately, as sample days (p12502, 13-4), and included in the calculation of the nucleation frequency (p12501, 125).

**Response:** *Thanks. In the revised manuscript, undefined sampling days were considered separately. The statistical results will be shown in the new Table 2 and Fig. 8. Only 11 sampling days were defined as undefined days.*

P12502, 16-15: The seasonal variation of the nucleation frequency which is observed at the SORPES station, with higher frequencies in spring and summer, has already been reported several times, and is typically explained by higher radiation and stronger biogenic activity (Manninen et al., 2010).

The statement “Such fronts were generally not strong enough to improve the air quality in Nanjing” suggests that in winter, low nucleation frequencies at the SORPES site could be mainly explained by higher CS. It is true that at boundary layer stations, higher CS are more frequently found on nonevent days compared to event days (Manninen et al., 2010), but based on Table 1 it seems that at the SORPES station the CS does not show seasonal variations which are significant enough to drive the seasonal pattern of the nucleation frequency. Thus I would remove the statement, and only discuss the influence of the CS/PM<sub>2.5</sub> in section 3.2.1, together with the other atmospheric parameters.

**Response:** *Thanks. In the revised manuscript, we emphasized the similar seasonal variation of NPF frequency with other boundary layer sites and referred the article (Manninen et al, 2010). We omitted the sentence “Such fronts were generally not strong enough to improve the air quality in Nanjing” and discuss the PM<sub>2.5</sub>/CS in section 3.2.1.*

p12503, 11: Why did you choose to work on the size range 6-30 nm instead of 7-20 nm, which is

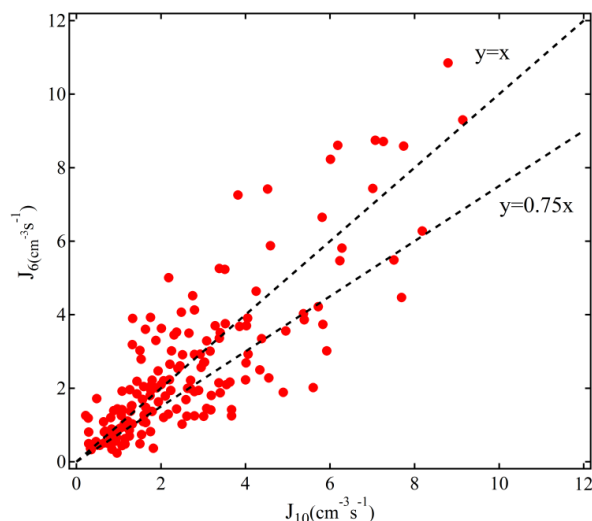
more commonly used (Hirsikko et al., 2007)?

**Response:** *Growth rate in the size range 7-20 nm are often calculated for the ions data (e.g. AIS data) because this size range is usually defined as 'Large ions'. In this study, we use the DMPS data and 6-30 nm is defined as 'nucleation mode'. In addition, as the growth rate is quite high at SORPES (Hermann et al., 2014), a large size range is helpful to decrease the uncertainty (Vakkari et al., 2011).*

p12503, 17-11: Given the ranges which are reported for the formation rates at other Chinese stations, it is quite complex to accurately state if the formation rates from the SORPES site are comparable to other measurements; average values would help.

Moreover, since the formation rate has already been reported to decrease with size because of the coagulation process (e.g. Kulmala et al., 2013), I wonder if the comparison of J3, J5.5, J6 and J10 is relevant.

**Response:** *Thanks. In the revised manuscript we will provide the average values from the campaign / sites except those with averaged values were not available. It is the fact that the size range of formation rate is different in each work due to different cut-off of instrument (6 nm in this work). The below figure suggests that sometimes the  $J_{10}$  was higher than  $J_6$ , which may be due to the instrument issue (e.g. loss the small particles, see the Appendix) or the stronger NPF occurred at high altitude and mixed into sampling height (Yli-Juuti et al., 2011). Event through, a comparison, a comparison is still useful to understand the general difference or similarity.*



p12503, 118-20: I would suggest to precise that this result is typically observed at other boundary layer stations (e.g. Manninen et al., 2010).

**Response:** *Thanks. We will make a change in the revised manuscript.*

*The discussion about CS was moved to section 3.2.2 and the values of CS in Table 2 were deleted.*

*In Figure 9 we show the CS during the event days and non-event days.*

p12503-12504, section 3.2.2: NPF seems to be favored by lower RH values at the SORPES, but at the same time high NPF frequencies are reported in summer, when rainfall is maximum. Can you please comment this observation? How do the rain have a diurnal pattern?

**Response:** *The maximum of total rainfall amount in summer is due to more heavy rain related to the Asian summer monsoon. A high NPF frequencies in summer is a result of higher solar radiation, oxidation capacity of the atmosphere, and also a low CS. Rain in this region has a diurnal pattern as more rain in later afternoon and evening when the convection are well-developed. We will add some discussions to address this point in the revised manuscript.*

Several atmospheric parameters, such as radiation, ozone, SO<sub>2</sub>, humidity and PM<sub>2.5</sub> are discussed separately. Such analysis are often performed, despite the fact that all these parameters probably have combine effects. In the present study, an additional calculation of a proxy for the sulphuric

acid concentration (e.g.: Petaja et al., 2009, Mikkonen et al., 2011) would permit to simultaneously consider the influence of several parameters on the occurrence of NPF.

I would also include backward trajectories in the analysis and discuss the nucleation frequencies associated to the different clusters, which might be related to different gaseous precursors. In fact, the discussion is focussed on SO<sub>2</sub>, and based on Fig. 6, other vapours (VOCs) should, at least, be mentioned.

**Response:** *Thanks. We will calculate the sulphuric acid proxy based on the equation described by Mikkonen et al (2011). We used the 'Atlantic equation' which performed better at SORPES (Hermann et al., 2014). Two sub-figures will be added (CS and [H<sub>2</sub>SO<sub>4</sub>]). We will also add a Table to discuss the NPF event in different air masses.*

p12503, 126: Considering the large variability of the measurements, I would remove the word “significantly”.

**Response:** *Thanks. We will make a correction.*

p12504, 11-4: I think that some references are needed to discuss the roles of humidity and temperatures, which can be quite complex. Some studies, such as the one from Young et al. (2007), suggest that NPF could be favored by low temperatures, which contrasts the results of the present work. Moreover, if low RH has already been reported to favor the occurrence of NPF (Birmili et al., 2003) and to promote higher cluster concentrations and nucleation rates (Jeong et al., 2004, Sihto et al., 2006), nucleation events have also been observed in the vicinity of clouds, where high RH are found (Clarke et al., 1998).

**Response:** *Thanks. In revised manuscript some articles were referred to discuss the roles of air temperature and RH.*

p12504, 19: Regarding the variability on Fig. 9f, I think again that the conclusions should be more balanced, which SO<sub>2</sub> concentrations being “on average” higher on event days.



***Response:*** Thanks. We will make a correction.

p12505, 11-3: This result is not surprising and only reflects the fact that different processes and vapours are involved in 1) the formation of the clusters and 2) the growth of these clusters. This observation was previously reported by studies conducted in different environments (e.g., Yli-Juuti et al., 2011, Rose et al., 2015).

***Response:*** Thanks. We will add some discussions accordingly.

p12505, 14-7: If an anti-correlation between J and RH is reliable (and has already been observed, e.g. Sihto et al., 2006), the correlation between GR and RH itself is likely to be an artefact, and could rather indicate a correlation between GR and other parameters which share the same origin as RH. This assumption is supported by 114-16: higher GR are found in air masses passing over the polluted YRD area, which are certainly characterised by high humidities, but also by a large pool of vapours (anthropogenic VOCs?) which might be involved in the growth process.

***Response:*** Thanks. We will add some discussions accordingly.

p12505, 117-20: I would suggest to move this sentence to section 3.2.2.

***Response:*** Thanks. We will move it to section 3.2.2.

p12506: I would add temperature and relative humidity to Fig. 12, and to highlight the differences between 2012 and 2013, I would show the same figure for 2012.

***Response:*** We will add temperature and RH in Fig. 12 and show the same figure for 2012.

Abstract/summary: should be modified according to the changes in the other sections of the manuscript.

**Response:** *We will revise the abstract/summary according to the changes.*

#### Other comments

In order to correct some minor grammatical errors (some of them are listed below), I would recommend that the manuscript is read by a native English speaker.

p12492, l28: “of” instead of “by”.

p12493, l8: Missing dot at the end of the line.

p12493, l12: Missing space in “Dal Maso”.

p12495, l12-14: The comma should be removed.

p12495, l22: “is measured by CPC”, instead of “are measured”.

p12495, l24: The word “about” should be removed.

p12495, l28: “were made” instead of “was made”.

P12496, l11 and l15: It should be *the* condensation sink and *the* coagulation sink.

P12498, l16: highest concentrations

p12499, l6: “burning” instead of “burnings”

p12501, l8: The word “as” should be removed.

p12503, l26: Check the Figure numbers

**Response:** *Thanks. We will revise the manuscript according to all these technical comments. The revised manuscript will be edited by an English native speaker.*

#### **Reference:**

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