Aerosol size distribution and new particle formation in western Yangtze River Delta of China: two-year measurement at the SORPES station

The paper by Qi et al. discusses measurements of submicron particles (6 – 800 nm) conducted between December 2011 and November 2013 at a suburban site of East China. Particle size distributions were described as a combination of three log-normal ditributions corresponding to the nucleation (6 - 30 nm), Aitken (30 – 100 nm) and accumulation (100 – 800 nm) modes. The seasonal and diurnal variations of the particle concentration in each mode are invastigated and the influence of air masses is discussed. The occurrence of NPF is also reported, and events are classified following the commonly used classification from Dal Maso et al, (2005), which includes two main classes. Some additional information (formation and growth rates) are also provided for the strongest events, belonging to class I. Several atmospheric parameters which are typically found to influence NPF, including temperature, relative humidity, radiation, SO_2 and the condensation sink are considered in the analysis. Finally the end of the paper focusses on high nucleation frequencies observed in August 2013.

I recommend the publication of this paper as it provides new measurements of aerosol size distributions and observations of NPF in China from a 2-year dataset, which has rarely been done before. Such long term observations are particularly of great interest to improve our knowledge of the NPF process, which remains incomplete. However, I have several comments and recommandations which are described in the following and should be addressed in a revised version.

Specific comments

p12496, l18: The hygroscopic growth of particles has been shown to have a significant effect on sink calculations (e.g. Hõrrak 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.

p12497, l19-21: If particle concentrations from the urban site near Beijing (32 700 cm^{-3}) are considered comparable to those measured at the SORPES station (19200 ± 9200 cm^{-3}), concentrations from the rural site (11500 cm^{-3}) should also be considered as similar, and not lower.

p12497, l25: 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^{-3} are reported for the accumulation mode at the SORPES station, especially during summer (5300 ± 4200 cm^{-3}).

p12498, l1: Which concentration is correct: 5700 cm^{-3} from the text or 5800 cm^{-3} from Table 1?

p12498, l3: The definition of the seasons can slighlty 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.

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.

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.

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

p12498, l1-2: Regarding the sentence: "Radiation connected with NPF events and local direct emissions 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?

p12499, l9-13: The sentence is quite long, and the brackets do not ease the reading. Maybe two shorter sentences could help!

p12499, l25-27: 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, l6.

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

p12500, l2: The fragment "typical seasons" sounds strange.

p12500, l18: 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 backtrajectories 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)).

p12501, l1-3: 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.

p12501, l4-11: First, I would remove the sentence "With low…daytime", since it is a general statement that does not highlight the analysis related to the inluence 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.

Finaly, I suggest to show the position of Nanjing on the maps (Fig. 6), so it would be easier to follow the explanation from l9 to l11.

p12501, l18: Why were undefined and non-event days considered all together? I think undefined days should be considered separately, as sample days (p12502, l3-4), and included in the calculation of the nucleation frequency (p12501, l25).

P12502, l6-15: The seasonal variation of the nucleation frequency which is observed at the SORPES station, with higher fequencies 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 non-event 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 2.5 in section 3.2.1, together with the other atmospheric parameters.

p12503, l1: 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)?

p12503, l7-11: 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 J_3, $J_{5.5}$, J_6 and J_{10} is relevant.

p12503, l18-20: I would suggest to prescise that this result is typically observed at other boundary layer stations (e.g. Manninen et al., 2010).

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?

Several atmospheric parameters, such as radiation, ozone, SO_2, humidity and PM2.5 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.: Petäjä 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_2, and based on Fig. 6, other vapours (VOCs) should, at least, be mentionned.

p12503, l26: Considering the large variability of the measurements, I would remove the word “significantly”.

p12504, l1-4: 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).

p12504, l9: Regarding the variabibilty on Fig. 9f, I think again that the conclusions should be more balanced, which SO_2 concentrations being “on average” higher on event days.

p12505, l1-3: 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).

p12505, l4-7: If an anti-correlation between J and RH is reliable (and has already ben 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 l14-16: higher GR are found in air masses passing over the polluted YRD area, wich are certainly caracterised by high humidities, but also by a large pool of vapours (anthropogenic VOCs?) which might be involved in the growth process.

p12505, l17-20: I would suggest to move this sentence 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.

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

Other comments

In order to correct some minor gramatical 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 coma 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.

References

Birmili, W., Berresheim, H., Plass-Dülmer, C., Elste, T., Gilge, S., Wiedensohler, A. and Uhrner, U.: The Hohenpeissenberg aerosol formation experiment (HAFEX): a long-term study including size-resolved aerosol, H2SO4, OH, and monoterpenes measurements, Atmos Chem Phys, 3(2), 361–376, doi:10.5194/acp-3-361-2003, 2003.

Clarke, A. D., Varner, J. L., Eisele, F., Mauldin, R. L., Tanner, D. and Litchy, M.: Particle production in the remote marine atmosphere: Cloud outflow and subsidence during ACE 1, J. Geophys. Res. Atmospheres, 103(D13), 16397–16409, doi:10.1029/97JD02987, 1998.

Hirsikko, A., Bergman, T., Laakso, L., Dal Maso, M., Riipinen, I., Hõrrak, U. and Kulmala, M.: Identification and classification of the formation of intermediate ions measured in boreal forest, Atmos Chem Phys, 7(1), 201–210, doi:10.5194/acp-7-201-2007, 2007.

Hõrrak, U., Aalto, P. P., Salm, J., Komsaare, K., Tammet, H., Mäkelä, J. M., Laakso, L. and Kulmala, M.: Variation and balance of positive air ion concentrations in a boreal forest, Atmos Chem Phys, 8(3), 655–675, doi:10.5194/acp-8-655-2008, 2008.

Jeong, C.-H., Hopke, P. K., Chalupa, D. and Utell, M.: Characteristics of Nucleation and Growth Events of Ultrafine Particles Measured in Rochester, NY, Environ. Sci. Technol., 38(7), 1933–1940, doi:10.1021/es034811p, 2004.

Kulmala, M., Kontkanen, J., Junninen, H., Lehtipalo, K., Manninen, H. E., Nieminen, T., Petäjä, T., Sipilä, M., Schobesberger, S., Rantala, P., Franchin, A., Jokinen, T., Järvinen, E., Äijälä, M., Kangasluoma, J., Hakala, J., Aalto, P. P., Paasonen, P., Mikkilä, J., Vanhanen, J., Aalto, J., Hakola, H., Makkonen, U., Ruuskanen, T., Mauldin, R. L., Duplissy, J., Vehkamäki, H., Bäck, J., Kortelainen, A., Riipinen, I., Kurtén, T., Johnston, M. V., Smith, J. N., Ehn, M., Mentel, T. F., Lehtinen, K. E. J., Laaksonen, A., Kerminen, V.-M. and Worsnop, D. R.: Direct Observations of Atmospheric Aerosol Nucleation, Science, 339(6122), 943–946, doi:10.1126/science.1227385, 2013.

Manninen, H. E., Nieminen, T., Asmi, E., Gagné, S., Häkkinen, S., Lehtipalo, K., Aalto, P., Vana, M., Mirme, A., Mirme, S., Hõrrak, U., Plass-Dülmer, C., Stange, G., Kiss, G., Hoffer, A., Törő, N., Moerman, M., Henzing, B., de Leeuw, G., Brinkenberg, M., Kouvarakis, G. N., Bougiatioti, A., Mihalopoulos, N., O'Dowd, C., Ceburnis, D., Arneth, A., Svenningsson, B., Swietlicki, E., Tarozzi, L., Decesari, S., Facchini, M. C., Birmili, W., Sonntag, A., Wiedensohler, A., Boulon, J., Sellegri, K., Laj, P., Gysel, M., Bukowiecki, N., Weingartner, E., Wehrle, G., Laaksonen, A., Hamed, A., Joutsensaari, J., Petäjä, T., Kerminen, V.-M. and Kulmala, M.: EUCAARI ion spectrometer measurements at 12 European sites – analysis of new particle formation events, Atmos Chem Phys, 10(16), 7907–7927, doi:10.5194/acp-10-7907-2010, 2010.

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P. and Lehtinen, K. E.: Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiala, Finland, Boreal Environ. Res., 10(5), 323, 2005.

Mikkonen, S., Romakkaniemi, S., Smith, J. N., Korhonen, H., Petäjä, T., Plass-Duelmer, C., Boy, M., McMurry, P. H., Lehtinen, K. E. J., Joutsensaari, J., Hamed, A., Mauldin III, R. L., Birmili, W., Spindler, G., Arnold, F., Kulmala, M. and Laaksonen, A.: A statistical proxy for sulphuric acid concentration, Atmos Chem Phys, 11(21), 11319–11334, doi:10.5194/acp-11-11319-2011, 2011.

Petäjä, T., Mauldin III, R. L., Kosciuch, E., McGrath, J., Nieminen, T., Paasonen, P., Boy, M., Adamov, A., Kotiaho, T. and Kulmala, M.: Sulfuric acid and OH concentrations in a boreal forest site, Atmos Chem Phys, 9(19), 7435–7448, doi:10.5194/acp-9-7435-2009, 2009.

Rose, C., Sellegri, K., Velarde, F., Moreno, I., Ramonet, M., Weinhold, K., Krejci, R., Andrade, M., Wiedensohler, A. and Laj, P.: Frequent nucleation events at the high altitude station of Chacaltaya (5240 m a.s.l.), Bolivia, Atmos. Environ., 102, 18–29, doi:10.1016/j.atmosenv.2014.11.015, 2015.

Sihto, S.-L., Kulmala, M., Kerminen, V.-M., Dal Maso, M., Petäjä, T., Riipinen, I., Korhonen, H., Arnold, F., Janson, R., Boy, M., Laaksonen, A. and Lehtinen, K. E. J.: Atmospheric sulphuric acid and aerosol formation: implications from atmospheric measurements for nucleation and early growth mechanisms, Atmos Chem Phys, 6(12), 4079–4091, doi:10.5194/acp-6-4079-2006, 2006.

Tunved, P., Nilsson, E. D., Hansson, H.-C., Ström, J., Kulmala, M., Aalto, P. and Viisanen, Y.: Aerosol characteristics of air masses in northern Europe: Influences of location, transport, sinks, and sources, J. Geophys. Res. Atmospheres 1984–2012, 110(D7) [online] Available from: http://onlinelibrary.wiley.com/doi/10.1029/2004JD005085/full (Accessed 27 February 2015), 2005.

Yli-Juuti, T., Nieminen, T., Hirsikko, A., Aalto, P. P., Asmi, E., Hõrrak, U., Manninen, H. E., Patokoski, J., Dal Maso, M., Petäjä, T., Rinne, J., Kulmala, M. and Riipinen, I.: Growth rates of nucleation mode particles in Hyytiälä during 2003−2009: variation with particle size, season, data analysis method and ambient conditions, Atmos Chem Phys, 11(24), 12865–12886, doi:10.5194/acp-11-12865-2011, 2011.

Young, L.-H., Benson, D. R., Montanaro, W. M., Lee, S.-H., Pan, L. L., Rogers, D. C., Jensen, J., Stith, J. L., Davis, C. A., Campos, T. L., Bowman, K. P., Cooper, W. A. and Lait, L. R.: Enhanced new particle formation observed in the northern midlatitude tropopause region, J. Geophys. Res. Atmospheres, 112(D10), D10218, doi:10.1029/2006JD008109, 2007.