

Dear anonymous reviewers,

Thank you very much for your comments on our manuscript [acp-2014-466]. Your comments and suggestions are valuable and very helpful for improving our manuscript. Based on your comments and suggestions, we carefully revised the manuscript, and the point-to-point responses to your comments and suggestions are listed below.

Thank you once again for your time and consideration.

Responds to the comments:

Note: All the revisions are based on the previously submitted manuscript in word format which corresponds to the ACPD. For the modified portions, we provide the page and line numbers of the ACPD format before modification.

Anonymous Referee #1

1. The paper should add more address clearly on time scale of averaging for meteorological factors, $PM_{2.5}$, CCN, BC as well as other data.

We thank you for your comments and suggestions. The time scales of averaging are 10 min for meteorological factors and chemical species, 5 min for $PM_{2.5}$, BC and atmospheric visibility, 4 min for aerosol size distributions and 1 hour for CCN. We have added this in the Figure

captions in our revised manuscript.

2. The authors should give more explanation for the equation (2).

We thank you for your comments and suggestions. More explanation and the derivation process for the equation 2 have been introduced in detail by Petters and Kreidenweis (2007) in ACP. Therefore, there is only brief summarization here.

The saturation (i.e. S) over an aqueous droplet can be calculated by:

$$S = a_{\omega} \exp\left(\frac{4\sigma_{s/a} M_{\omega}}{RT\rho_{\omega}D}\right)$$

According to the effect on the water activity of the solution, the hygroscopicity κ is defined as:

$$\frac{1}{a_{\omega}} = 1 + \kappa \frac{V_s}{V_{\omega}}$$

where V_s is the volume of the dry particulate matter and V_{ω} is the volume of the water.

For the equilibrium of a multicomponent system, the total volume of the water is the sum of the water contents due to the individual components, i.e. $V_{\omega} = \sum V_{\omega i}$ by using the ZSR (Zdanovskii, Stokes and Robinson) assumption. The individual $V_{\omega i}$ can be derived for $a_{\omega i} = a_{\omega}$.

$$V_{\omega} = \frac{a_{\omega}}{1 - a_{\omega}} \sum_i \kappa_i V_{s i}$$

The total volume of the system (water+solute) is

$$V_T = \sum_i V_{s i} + \sum_i V_{\omega i} = V_s + V_{\omega}$$

The individual dry component volume fractions are defined as $\varepsilon_i = V_{s i} / V_s$,

then

$$V_T - V_S = \frac{a_\omega}{1 - a_\omega} V_s \sum_i \kappa_i \varepsilon_i$$

Defining D_d as the dry diameter, $D_d^3 = 6V_S/\pi$, also $D^3 = 6V_T/\pi$

Then we can derive the equation 2 in ACPD, namely “ κ -Köhler theory”.

3. More related references should be added and the reference style should be adjusted correctly.

We thank you for your comments and suggestions. We have added more related references in correct style highlighted in red in our revised manuscript as following:

Dal Maso, M., Kulmala, M., Lehtinen, K. E. J., and Mäkelä J. M.:
Condensation and coagulation sinks and formation of nucleation mode particles in coastal and boreal forest boundary layers, *J. Geophys. Res.*, 107, doi:10.1029/2001JD001053, 2002.

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P., and Lehtinen, K. E. J.: Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland, *Boreal Env. Res.*, 10, 323-336, 2005.

Dal Maso, M., Sogacheva, L., Aalto, P. P., Riipinen, I., Komppula, M., Tunved, P., Korhonen, L., Suur-Uski, V., Hirsikko, A., Kurtén, T., Kerminen, V. -M., Lihavainen, H., Viisanen, Y., Hansson, H. -C., and Kulmala, M.: Aerosol size distribution measurements at four Nordic field stations: identification, analysis and trajectory analysis of new

- particle formation bursts, *Tellus*, 59B, 350-361, 2007.
- Fuchs, N. A. and Sutugin, A. G.: High-dispersed aerosols in *Topics in Current Aerosol Research*, edited by: Hidy, G. M. and Brock, J. R., Pergamon, Oxford, 2, 1-60, 1971.
- Gao, J., Chai, F. H., Wang, T., Wang, S. L., and Wang, W. X.: Particle number size distribution and new particle formation: New characteristics during the special pollution control period in Beijing, *J. Environ. Sci.*, 24, 14-21, 2012.
- Gong, Y. G., Hu, M., Cheng, Y. F., Su, H., Yue, D. L., Liu, F., Wiedensohler, A., Wang, Z. B., Kalesse, H., Liu, S., Wu, Z. J., Xiao, K. T., Mi, P. C., and Zhang, Y. H.: Competition of coagulation sink and source rate: New particle formation in the Pearl River Delta of China, *Atmos. Environ.*, 44, 3278-3285, 2010.
- Huang, Y. L., Li, L., Li, J. Y., Wang, X., Chen, H., Chen, J. M., Yang, X., Cross, D. S., Wang, H., Qiao, L. P., and Chen, H.: A case study of the highly time-resolved evolution of aerosol chemical and optical properties in urban Shanghai, China, *Atmos. Chem. Phys.*, 13, 3931-3944, doi:10.5194/acp-13-3931-2013, 2013.
- Hussein, T., Martikainen, J., Junninen, H., Sogacheva, L., Wagner, R., Dal Maso, M., Riipinen, I., Aalto, P. P., and Kulmala, M.: Observation of regional new particle formation in the urban atmosphere, *Tellus B*, 60, 609-521, doi:10.1111/j.1600-0889.2008.00365.x, 2008.

Kristensson, A., Dal Maso, M., Swietlicki, E., Hussein, T., Zhou, J., Kerminen, V. -M, and Kulmala, M.: Characterization of new particle formation events at a background site in Southern Sweden: relation to air mass history, *Tellus* 60B, 330-344, doi:10.1111/j.1600-0889.2008.00345.x, 2008.

Kumala, M., Dal Maso, M., Mäkelä J. M., Pirjola, L., Väkevä M., Aalto, P., Miikkulainen, P., Hämeri, K., and O'Dowd, C. D.: On the formation, growth and composition of nucleation mode particles, *Tellus B*, 53, 479-490, 2001.

Kulmala, M., Petäjä T., Mönkkönen, P., Koponen, I. K., Dal Maso, M., Aalto, P. P., Lehtinen, K. E. J., and Kerminen, V. -M.: On the growth of nucleation mode particles: source rates of condensable vapor in polluted and clean environments, *Atmos. Chem. Phys.*, 5, 409-416, doi:10.5194/acp-5-409-2005, 2005.

Shen, X. J., Sun, J. Y., Zhang, Y. M., Wehner, B., Nowak, A., Tuch, T., Zhang, X. C., Wang, T. T., Zhou, H. G., Zhang, X. L., Dong, F., Birmili, W., and Wiedensohler, A.: First long-term study of particle number size distributions and new particle formation events of regional aerosol in the North China Plain, *Atmos. Chem. Phys.*, 11, 1565-1580, doi:10.5194/acp-11-1565-2011, 2011.

Vehkamäki, H., Dal Maso, M., Hussein, T., Flannagan, R., Hyvärinen, A., Lauros, J., Merikanto, J., Mönkkönen, P., Pihlatie, M., Salminen, K.,

Sogacheva, L., Thum, T., Ruuskanen, T. M., Keronen, P., Aalto, P. P., Hari, P., Lehtinen, K. E. J., Rannik, Ü., and Kulmala, M.: Atmospheric particle formation events at Värriö measurement station in Finnish Lapland 1998-2002, *Atmos. Chem. Phys.*, 4, 2015-2023, doi:10.5194/acp-4-2015-2004, 2004.

Wang, X. F., Zhang, Y. P., Chen, H., Yang, X., and Chen, J. M.: Particle nitrate formation in a highly polluted urban area: a case study by single-particle mass spectrometry in Shanghai, *Environ. Sci. Technol.*, 43, 3061-3066, 2009.

Wang, Z. B., Hu, M., Sun, J. Y., Wu, Z. J., Yue, D. L., Shen, X. J., Zhang, Y. M., Pei, X. Y., Cheng, Y. F., and Wiedensohler, A.: Characteristics of regional new particle formation in urban and regional background environments in the North China Plain, *Atmos. Chem. Phys.*, 13, 12495-12506, 3013.

Wu, Z. J., Hu, M., Lin, P., Liu, S., Wehner, B., and Wiedensohler, A.: Particle number size distribution in the urban atmosphere of Beijing, China, *Atmos. Environ.*, 42, 7967-7980, doi:10.1016/j.atmosenv.2008.06.022, 2008.

4. I think there is misprint for the description of Figure7, please rewrite it.

We thank you for your good comments and valuable suggestions. We have rewritten the caption of Figure 7 as following:

Figure 7: Temporal evolutions of mode, median and geometric mean diameters for the entire aerosol size range and nanoparticle concentration (10-20 nm) on 3-4 April 2012.

5. The growth rate and formation rate calculation in this study is the “apparent particle formation and growth rate (i.e. APFR and APGR)”, However, I also see “FR” and “GR” in the text, please specify.

We thank you for your good comments and suggestions. We have specified them in our revised manuscript.

6. There are some spaces for improvement in English.

We thank you for your good comments. We have read the manuscript vary carefully and corrected any places having grammar errors or mistakes, which are highlighted in red in our revised manuscript.

Anonymous Referee #2

Specific comments:

1. The authors do not comment on the relatively large difference in the frequency of formation events in their springtime study (27%) with that observed during winter by Du et al. of 5.4%. This seasonal preference for greater frequency during spring is seen by many others in northern hemisphere locations and probably deserves to be pointed out.

We thank you for your comments and suggestions. Many studies have observed greater NPF frequency in springtime in northern hemisphere.

For example, seasonal NPF pattern with a spring maximum and winter minimum is typical for all Nordic stations (Dal Maso et al., 2007; Kristensson et al., 2008; Vehkamäki et al., 2004). In North China Plain, The number of events was highest in the spring months (Wang et al., 2013).

The high frequency during spring in urban Shanghai is probably due to high frequency of strong wind from northern China, which helps removing the pre-existing particles in the atmosphere and further favors the occurrence of new particle formation events (Wu et al., 2008; Wang et al., 2013). We have added the analysis in our revised manuscript, which is highlighted in red.

2. The instrumentation descriptions are quite terse. For particle size distributions, the mention of TSI 3080 is insufficient information. What was the CPC? Was the TSI software used? If so, was multiple charge and diffusion correction applied? What was the sample inlet? Were diffusion losses and impaction losses in sample lines accounted for? If all this information is in a previous publication, please cite that paper.

We thank you for your good comments and suggestions. The SMPS data are recorded by AIM (Aerosol Instrument Management) software from TSI company. The SMPS 3936 (TSI corp.) is employed to track the size distribution change, in which the CPC 3736 (TSI corp.) is used to count the number of particle of each size. The neutralizer 3077a (TSI

corp.) is used in the system to provide known charge on the particles going into the SMPS. The size of the employed impactor is 0.071 cm. Both multiple charge and the diffusion correction is applied. The inlet information has been reported in our previously papers (Wang et al., 2009; Huang et al., 2013).

Wang, X. F., Zhang, Y. P., Chen, H., Yang, X., and Chen, J. M.: Particle nitrate formation in a highly polluted urban area: a case study by single-particle mass spectrometry in Shanghai, *Environ. Sci. Technol.*, 43, 3061-3066, 2009.

Huang, Y. L., Li, L., Li, J. Y., Wang, X., Chen, H., Chen, J. M., Yang, X., Cross, D. S., Wang, H., Qiao, L. P., and Chen, H.: A case study of the highly time-resolved evolution of aerosol chemical and optical properties in urban Shanghai, China, *Atmos. Chem. Phys.*, 13, 3931-3944, 2013.

3. Similarly, the Thermo FH62C14 needs another line of two of description. It should be included in the paper that this is a beta attenuation gauge. Also, there should be some mention of QA procedures, the detection limit, etc. Once again, if this is included in a previous paper, please cite the paper. Finally along these lines, the model number of the Vaisala visibility monitor should be included.

We thank you for your good comments and suggestions. The Thermo FH62C14 Continuous Ambient Particulate Monitor (FH62C14) is a

radiometric particulate mass monitor capable of providing real-time measurements. It incorporates time-averaged measurements of an integral beta attenuation sensor and advanced firmware to optimize the continuous mass measurement. The FH62C14 equips a dynamic heating system (DHS) to maintain the relative humidity (RH) of the air passing through the filter tape of the radiometric stage well below the point at which the collected particles accrete and retain liquid water. The DHS system minimizes the internal temperature rise ensuring negligible loss of semi-volatiles from the collected sample when the ambient RH is below the threshold to which the heater is controlling. As the ambient RH increases above the threshold, the applied heating is optimized to maintain the RH threshold above the beta attenuation filter tape. Necessary sensor calibrations are performed for temperature, relative humidity, barometric pressure and volumetric flow regularly to maintain valid measurements.

We have added this description in our revised manuscript which is highlighted in red.

4. The authors mention the condensation sink in Section 3.2.2 and describe a method for theoretical estimation of particle formation rate. Using the method of Dal Maso et al (2002, JGR) the author could estimate the numerical values of the condensation (and coagulation?) sinks and strengthen their analysis.

We thank you for your comments and valuable suggestions. We have added the comparison of condensation sink between the NPF event days and non-event days to strengthen our analysis. The added parts are highlighted in red in our revised manuscript.

5. The discussion in Section 3.2.4 describes the method for determining an estimated kappa value for the measurement period. The work seems quite good, but a detailed reading reveals a number of unanswered questions: 1) Are all the data for this section limited to the 70 hours used for the graph in Figure 10? (line 25); 2) If only 70 hours of data were used, why was only this small subset of data chosen, and how was it chosen? 3) What was the effective kappa used for the predicted N(CCN)? How does it compare to other estimates of this parameter? (There does seem to be sufficient information for the reader to calculate the effective kappa, but it just seems strange that the authors do not present this useful result!)

In this paper, we examined the closure study during NPF events. Due to MARGA data limitations, we only attempted to get CCN closure for the case NPF event in this study (i.e. 10:00 on 3 to 4:00 on 4 April, 18 hours in total). The predicted CCN concentrations were compared with the measured ones under five SS level (i.e. 0.2%, 0.4%, 0.6%, 0.8% and 1.0%), therefore 90 (not 70, we are very sorry for neglecting the typewriting error during the proofreading stage) groups of data were

presented in Figure 10.

Correspondingly, the 1 h mean effective kappa values are from 0.19 to 0.42, and average at 0.28 during the case NPF event. The effective kappa values were used to calculate the critical dry diameter, by which we can predict CCN number concentrations through integration aerosol number concentrations between the bottom and top critical dry diameters.

We have added the analysis which is highlighted in red in our revised manuscript.

Technical Corrections:

1. p: 18644, line 5-6: A reference is given in the text as Zhang et al., 2010. In the references section there is a Zhang, 2010; and Zhang et al., 2012 and 2013. Please clarify.

We thank you for your comments and kind remind. In p: 18644, line 5-6, the reference given in the text is Zhang, 2010. We have corrected this in our revised manuscript.

2. p: 18644, line 22-24: the clause that begins” and atmospheric ammonia” is awkward and difficult to make sense of. Please reword-split into two sentences if necessary.

We thank you for your comments and suggestions. The sentence has been reworded in our revised manuscript as following:

“...Atmospheric ammonia can effectively lower the surface pressure of gaseous sulfuric acid molecular and participate homogeneous

nucleation with gaseous sulfur acid and water vapor (Smith et al., 2004; Sakurai et al., 2005; Gaydos et al., 2005).”

The corrections are highlighted in red in our revised manuscript.