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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Insight into winter haze formation mechanisms based on aerosol hygroscopicity and effective density measurements" *by* Yuanyuan Xie et al.

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Anonymous Referee #1 Received and published: 8 March 2017 The authors presented comprehensive aerosol dataset observed from metropolitan city of Shanghai. The measurements and data are valuable to study nowadays severe haze in China. The authors conclude that the accumulation of local emissions under stagnant meteorological conditions as well as rapid particle growth by secondary processes are primarily responsible for the haze formation in Shanghai. The analysis of particles hygroscopisity and density variations during pollution events is very interesting although no specific mechanism, which is actually very complex in urban areas, is addressed in the study. And also, the authors may need to improve the language. In general, I think the paper

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Discussion paper

is suitable for publication in this special issue after addressing some minor issues as follows,

AnswerïijŽWe sincerely thanks you for your pertinent comments and valuable suggestions. The language has been polished in the revised manuscript.

L36 remove “in heavily polluted areas” .

Answer: Revised.

L88 no mechanisms are actually discussed in this paper.

Answer: The banana-shaped particle size distribution provide a unique chance to reveal evolutions of particle hygroscopicity and effective density due to particle growth.

Section 2.1, Besides the sampling sites information, the authors also present measurements and data information here.

Answer: Revised. Measurements and data information in this section are categorized in another section.

L172 the authors think that the differences among the concentrations of PM1, PM2.5 and PM10 were insignificant. Is that true? According to the Fig. 2, on 26 Dec, they showed large differences in PM1, PM2.5 and PM10.

Answer: The statement has been revised as “Generally, the difference between the concentrations of PM1.0 and PM2.5 during clean periods was less significant than that in haze days”.

L176 -177, you mentioned that the PM mass dropped sharply due to the atmospheric dilution or precipitation. Do you have such data to support this?

Answer: This conclusion is support by meteorological data in the revised Figure S1. Detailed description has been added: During the end of each PM episode, the change in weather conditions played a key role in the decrease of particle concentration. As

shown in Figure S1, the prevailing winds on haze days were from the northwest. The prevailing winds during two clean periods (December 25-27 and January 12-14) were northeasterly, which bring clean air mass from East China Sea. Two cold fronts from the north swept Shanghai on December 31 and January 6, bringing gale and lower temperature, which favored the dispersion of atmospheric pollutants.

L182, is it 0.28, or 0?

Answer: it is 0.28

L202, it seems particles with Dp>300 nm are with lower kappa, why? Some explanations are needed here.

Answer: For 300-400 nm particles, the average Kappa are similar (0.335 for 300 nm, 0.331 for 350 nm, and 0.335 for 400 nm), whereas the 5th percentile ïĄń decreased with increasing size. Additional statement is given as "It is noticeable that the 5th percentile hygroscopicity decreased for dry diameter larger than 300 nm, likely due to the presence of the smallest dust particles (Gasparini et al., 2006)".

L217ïijŽ The interpretation ". . .strong formation of sulfate and nitrate" looks contrary with the section 3.2, the section 3.2 shown that SNA (sulfate, nitrate, ammonium) only accounts for 28% of PM1.0.

Answer: The formation of sulfate and nitrate is stronger compared to the USA site reported by Gasparini et al. (2006). The statement has been revised as "We attribute the different size dependencies of hygroscopicity among various measurement sites to the total emissions of SO2 and NOx, which were responsible for the formation of hygroscopic sulfate and nitrate".

L235-238: The reviewer is confused that why the number fraction of the lower density group increased as the concentration of NO increase. Did the authors analyze the relationship of them? or any reference?

Answer: This feature was reported in two papers. The statement has been revised

as: The lower density particles with ïĄšeff < 1.0 g cm-3 were attributable to fresh or partially aged traffic-related particles, because the number fraction of the lower density group in urban area was found to be consistent with the concentration of NO (indicator of traffic) (Levy et al., 2013;Rissler et al., 2014).

Section 3.3, you talk about Kappa in the first part of this section, but you used GF in the second part. It'd better to use one parameter.

Answer: The term "GF" in the second part is replaced by "Kappa" in the revised manuscript.

Fig.3 the authors may look the mass fraction of SIA, but not the mass concentrations. It is of course that the mass of each component will increase with the increase of PM.

Answer: The mass fraction of SIA is clearly reflected by linear regressions.

Fig.5 and L259-261, it seems it's difficult to see the characteristics you described here. You may replot the figure to make it more clearly to reviewers.

Answer: The particle number concentrations (black line in Figure 5) during haze period varied in the same range as in transition period, indicating little difference. The volume concentration (purple line) in haze days was always higher than that during transition period.

L342-344, is the first banana shape a NPF event? Because you said the other two are not.

Answer: The possibility of NPF can be ignored in this observation due to the absence of the burst of nucleation mode particles. Difference between NPF and the three particle growth events has been discussed in detail in the revised manuscript.

L360-371: The science of the analyzing method is weak. Anytime the number fraction and GF of the more-hygroscopic group always increase with particle size (Figure 6). It's hard to say that the hygroscopicity difference in size was caused by the particle

growth. The density difference has also the similar problem.

Answer: Indeed, this feature that the GF of the more-hygroscopic group increase with particle size cannot be attributed to particle growth in most cases, because the particles in different size are very likely from different source. In this observation, particle growth process was clearly displayed by the banana-shaped evolutions of particle size distribution, which provided a unique chance for us to study hygroscopicity evolution due to particle growth.

Section 3.6, it's very interesting to look at the particles hygroscopisity and density evolutions during the particle growth. The authors have investigated the particles with different Dp (40 nm, 100 nm and 220 nm). But to my understand, it may be more reasonable to look the GF and density with same Dp during different stage. For example, how do the GF and density of 100 nm particles changed from initial stage to growth stage?

Answer: Temporal variation of GF for a certain size was extensively discussed in previous studies. Different from most studies, one highlight of this work is the particle growth process reflected by the "banana" shape particle size distribution. The objective is to reflect the changes in hygroscopicity and effective density as particle is growing. This statement has been added: The latter two banana-shaped evolutions lasted long enough to tracer the changes in hygroscopicity and effective density due to particle growth.

And also, according to Fig. 7, it seems, during the period 2 and period 3, the concentrations for both the Nitrate and sulfate didn't increase and remain flat trends. How can you say that the secondary sulfate and nitrate was major contributors to particle growth during haze events?

Answer: The relative contribution of nitrate determined by the SPAMS (0.2ïĂ■2.0 ïĄ■m) increased visibly as the PM episode developed. Different from the total concentration in SPAMS, the HTDMA test showed that the hygroscopcity increased as the particle grew

from 40 nm to 100 nm, revealing hygroscopic SNA contributed greatly to the particle growth from 40 nm to 100 nm particles.

Reference Gasparini, R., Li, R. J., Collins, D. R., Ferrare, R. A., and Brackett, V. G.: Application of aerosol hygroscopicity measured at the Atmospheric Radiation Measurement Program's Southern Great Plains site to examine composition and evolution, J. Geophys. Res.-Atmos., 111, D05S12, doi:10.1029/2004JD005448, 10.1029/2004jd005448, 2006. Levy, M. E., Zhang, R. Y., Khalizov, A. F., Zheng, J., Collins, D. R., Glen, C. R., Wang, Y., Yu, X. Y., Luke, W., Jayne, J. T., and Olaguer, E.: Measurements of submicron aerosols in Houston, Texas during the 2009 SHARP field campaign, J. Geophys. Res.-Atmos., 118, 10518-10534, 10.1002/jgrd.50785, 2013. Rissler, J., Nordin, E. Z., Eriksson, A. C., Nilsson, P. T., Frosch, M., Sporre, M. K., Wierzbicka, A., Svenningsson, B., Londahl, J., Messing, M. E., Sjogren, S., Hemmingsen, J. G., Loft, S., Pagels, J. H., and Swietlicki, E.: Effective density and mixing state of aerosol particles in a near-traffic urban environment, Environ Sci Technol, 48, 6300-6308, 10.1021/es5000353, 2014. Gasparini, R., Li, R. J., Collins, D. R., Ferrare, R. A., and Brackett, V. G.: Application of aerosol hygroscopicity measured at the Atmospheric Radiation Measurement Program's Southern Great Plains site to examine composition and evolution, J. Geophys. Res.-Atmos., 111, D05S12, doi:10.1029/2004JD005448, 10.1029/2004jd005448, 2006. Gasparini, R., Li, R. J., Collins, D. R., Ferrare, R. A., and Brackett, V. G.: Application of aerosol hygroscopicity measured at the Atmospheric Radiation Measurement Program's Southern Great Plains site to examine composition and evolution, J. Geophys. Res.-Atmos., 111, D05S12, doi:10.1029/2004JD005448, 10.1029/2004jd005448, 2006. Gasparini, R., Li, R. J., Collins, D. R., Ferrare, R. A., and Brackett, V. G.: Application of aerosol hygroscopicity measured at the Atmospheric Radiation Measurement Program's Southern Great Plains site to examine composition and evolution, J. Geophys. Res.-Atmos., 111, D05S12, doi:10.1029/2004JD005448, 10.1029/2004jd005448, 2006.

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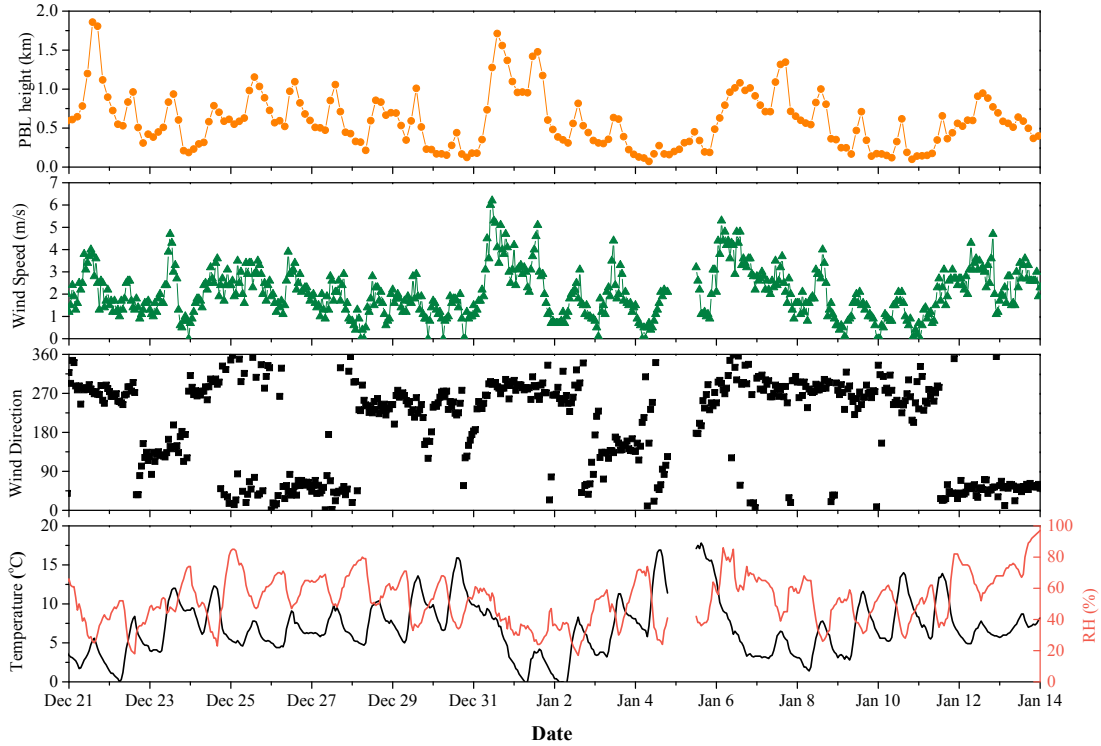

Fig. 1. Figure S1

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