

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

**Anonymous Referee #2**

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In this study, aerosol measurements were performed over about three weeks during winter to understand the causes of severe haze pollution in Shanghai. The measured aerosol properties include particle size distributions, hygroscopicity, effective density, and chemical composition. From the analysis of aerosols, trace gases, and meteorological data, it is concluded that the particle pollution events are caused by the accumulation of local emissions under stagnant meteorological conditions and exacerbated by rapid particle growth via secondary processes. Overall, the study is well executed, data analysis is mostly appropriate, and the paper is reasonably well written. I believe that it would be beneficial to extend the analysis to include several other effects, as detailed below. Also, a number of minor issues need to be addressed before the paper

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can be accepted for publication.

A recent publication by Wang, G., et al. (Persistent sulfate formation from London Fog to Chinese haze. Proc. Natl. Acad. Sci. USA 2016, 113 (48), 13630-13635) has shown that in two other major Chinese cities the aqueous oxidation of SO<sub>2</sub> by NO<sub>2</sub> in the absence of light can lead to efficient sulfate formation on fine aerosols. The process requires high relative humidity and the presence of NH<sub>3</sub>. It is suggested that in heavily polluted environments, this heterogeneous process can form large amounts of particulate sulfate and nitrate in aqueous particles. Do you have photoactinic light intensity measurements to evaluate the relative contributions from photochemical and dark reactions leading to the particle growth? Were ammonia measurements available for the study period? Can you use particle hygroscopicity measurements reported in your study to derive aerosol state (aqueous/dry) and relate with the particle growth rates? Doing so would bring this study to an entirely new level.

The authors should at least attempt to explain the 5-day cycle. Was it related to the workweek/weekend cycle or something else?

Minor comments:

L11: Particulate matter (PM) and haze are not synonymous, strictly speaking. The former term is typically used to describe aqueous aerosol particles (deliquesced, but not cloud droplets). These two terms cannot be interchanged; such use creates confusion. I suggest revising the use of haze and PM in the abstract and throughout entire manuscript.

L15: This sentence may become clearer if re-written as follows: “The mass ratio of SNA/PM<sub>1.0</sub> (sulfate, nitrate, and ammonium) fluctuated only slightly around 0.28, suggesting that both secondary inorganic compounds and carbonaceous aerosols contributed substantially to the haze formation, regardless of pollution level.” Also, the original sentence implies that all of the non-SNA material is carbonaceous. Perhaps this must be stated explicitly.

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L77: This statement implies that all traffic particles are soot aggregates, which is not correct

L78: Do the authors refer to material density or effective density?

L85: Must be 'cascade impactor' here and throughout the rest of the manuscript

L87: Mass spectrometry is used to measure the particle composition, which is used to infer the particle hygroscopicity and density.

L112: HTDMA does not measure the particle number size distribution

L132: '...Mass SpectrometER'

L166: these values must be rounded off, e.g., 57 +/- 37

L175: what does 'late' refer to?

L188: This sentence is confusing because it compares the contribution from a chemical (NO<sub>x</sub>) with that from a source of a chemical (presumably SO<sub>2</sub>) – coal-fired power plants. Also, doesn't coal combustion release NO<sub>x</sub> as well? The authors must provide data showing that traffic contributes more to the NO<sub>x</sub> burden than the power plants and other industrial sources that utilize coal.

L194: what does 'their' refer to?

L195: Isn't sulfate also of secondary origin?

L209: The meaning of this sentence is unclear. Why was hygroscopicity limited to smaller sizes? Do you mean 'measurements were limited to sizes smaller than 250 nm'?

L226: replace 'contradictory' with 'opposite'

L240: Insert a reference to Figure 2 early on in this paragraph

L282: Not all VOCs react with ozone. Can you provide data on the concentration of

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unsaturated organics?

L286: '...were less- and some that were more' - what?

L304 and several other instances: 'less-massive' – did you mean 'lower density'?

L381: '...contributed substantially...because the...ratio was almost constant...' – this is an invalid argument. The second part does not follow from the first part.

Figure 2: explain in figure caption the meaning of the dashed line

Figure 3: What is 'SIA' in figure legend. Use a secondary Y-axis for the SIA/PM ratio

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