

General comments:

This paper characterizes the aerosol physical, chemical, and optical properties and implication of visibility during an intense haze episode in Beijing winter. Heavy haze episodes occur more frequently in Beijing and the Great North China Plain. The manuscript provides numerous information on aerosol physicochemical properties and the effect on visibility due to these haze particulates. It is useful to quantitatively estimate the aerosol direct radiative forcing under heavy haze cases. Obviously, there are numerous grammatical and technical errors in the manuscript. This paper is reconsidered to be acceptable and published after major revisions.

Specific comments:

(1) Abstract, lines 20-24: “Light scattering apportionment showed that organic.....contributed to light scattering fractions of 57%....., respectively. This study indicated that the organic component in submicron aerosol plays an important role in visibility degradation in this haze episode in and around Beijing”.

⇒ As is well known that, both of scattering and absorption of aerosol particles can degrade the visibility. Many studies also found that urban aerosols under haze events in Beijing did have moderate absorption of solar light, which means that the absorption of aerosols may contribute to the visibility degradation. Therefore, to strongly support this conclusion, the authors should provide the visibility data versus organic compound during the haze episode in the context.

(2) Introduction, lines 13-14: “..., though the total aerosol mass concentration has decreased in Beijing in the last ten years.”

⇒ What does the “total aerosol mass concentration” mean? Total suspended particulate (TSP), PM₁₀ or the others? Please make clarify and add the citation of publications.

(3) Results and discussion, Page 23381, Lines 18-19: “Particularly at 08:00 of 28

January (Beijing time), the lapse ratio of temperature is nearly $-6^{\circ}/100\text{m}$, which means a very stable synoptic condition”.

⇒ This sentence makes contradictory with the Figure 3. Generally, the lapse rate of air temperature is about $-6^{\circ}\text{C}/100\text{m}$ on the global continent.

(4) Pages 23380-23381, Lines 25-26: “..., indicating that PM_1 dominated the total mass of aerosol particles”. And in Page 23383, Lines 9-10: “..., which indicated a more dominant coarse particle mode compared with the other locations.”

⇒ The two conclusions are contradictory and would confuse the readers. Please clarify it.

(5) Page 23384, Lines 5-6: “..., which indicates an increasing fraction of relative coarse aerosol, consistent with the variation pattern of $\text{PM}_1/\text{PM}_{2.5}$ showed in Fig. 1b.”

⇒ Please give a short interpretation about this conclusion.

Minor comments:

(1) Title: “Aerosol physicochemical properties and implication for visibility during an intense haze episode during winter in Beijing”

⇒ Change to “Aerosol physicochemical properties and implication of visibility during an intense haze episode in Beijing winter”.

(2)Page 23375, the affiliation of the authors: “Chinese Academy of Science”

⇒ Change to “Chinese Academy of Sciences”

Change “College of Atmospheric Science” to “College of Atmospheric Sciences”

(3)Abstract, line 3: change “during an extreme haze episode in Beijing” to “during an extremely intense haze episode in Beijing”

(4) Symbols such as PM_1 , $\text{PM}_{2.5}$ and PM_{10} in the abstract are not explained.

(5) Abstract, line 7: change “during the most heavily polluted periods” to “during the most heavily polluted period”

- (6) Abstract, lines 7-8: “The average scattering coefficient at 550 nm was $877 \pm 624 \text{ Mm}^{-1}$ ”
- (7) Abstract, line 12: “accumulation mode”
- (8) Abstract, line 14: “the mass concentrations of”
- (9) Abstract, line 16: “contributed greatly to the growth of particles during the heavily polluted day (28 January)”
- (10) Abstract, lines 17-20: “Increasing relative humidity and stable synoptic condition combined with heavy pollution on 28 January, led to enhanced water uptake by the hygroscopic submicron particles and formation of secondary aerosols, which might be the main reasons for the severity of the haze episode.”
- (11) Abstract, lines 22-24: “This study indicated that the organic component in submicron aerosol played an important role in visibility degradation during the haze episode in Beijing.”
- (12) Introduction, line 2: “Atmospheric aerosol particles play an significant role in...”
- (13) Introduction, line 4: “Ramanathan et al., 2001). In addition, they can act as...”
- (14) Introduction, line 6: “clouds are indirectly influenced by aerosols...”
- (15) Introduction, line 8: “due to its deleterious effect...”
- (16) Introduction, lines 12-14: “...has suffered substantially from the deterioration of air quality and the degradation of visibility, though the total aerosol mass concentration has decreased in the last ten years (**please add the citation**).”
- (17) Introduction, lines 16-20: “The frequency of visibility between 2 km and 10 km has increased from 37% in 1999 to 43% in 2007 (Zhang et al., 2010). The mass loading of fine aerosol particles and their precursors (e.g., NH_3 , volatile organic compounds (VOCs), SO_2 and NO_x)...and strong temperature inversion (Zhang et al., 2013).”
- (18) Introduction, lines 21-23: “In the past decades, many researches have been done to characterize the chemical and physical properties of aerosol particles in Beijing and its surrounding regions. These studies mainly focused on the following aspects:”
- (19) Introduction, line 28: The symbol of MODIS is not explained.
- (20) Page 23378, lines 8-14: “The above mentioned studies, based on either long-term

or short-term observations provided us with comprehensive knowledge of aerosol properties with near average....However, only a few studies were carried out under highly polluted days, and these studies mainly focused on....boundary layer dynamics (Huang et al.,). The interaction between chemical and physical properties of aerosol was seldom...”

(21) Page 23378, lines 17-18: “during pollution episodes.”

(22) Page 23378, line 20: Change “averaged” to “average”, and modify the other place in the manuscript.

(23) Page 23378, lines 21-26: “which was theas we know. In this study, we investigated the evolution of physical, chemical, and optical properties of urban aerosol particles during the haze episode by using the in-situ measurements.”

(24) Page 23379, line 2: “2.1 Site information and instrumentation”

(25) Page 23379, lines 3-5: “The aerosol sampling site was situated on the roof (about 15 m height above the surface) of ..., Chinese Academy of Sciences, which was located between...”

(26) Page 23379, lines 9-11: “angles between 7-170°) of low relative humidity (RH) aerosol at wavelengths of 450, 550 and 700 nm, respectively, without size-selective inlet. The nephelometer was operated at 5 Lmin⁻¹ with time resolution of 1 minute.”

According to (Anderson and Ogren, 1998), the correction factors of Angular Nonidealities for the nephelometer without size-selective inlet should be considered at different wavelengths, please add the discussion in the manuscript.

(27) Page 23379, line 14: No reference of“(Anderson and Ogren, 1998)” is found.

(28) Page 23379, line 20: “between 14 nm and 2500 nm”

(29) Page 23379, line 21: “comprising of ...”

(30) Page 23379, line 24: “The SMPS data covered the particle sizes range from 14 to 533 nm”

(31) Page 23379, lines 25-26: Please give a short discussion about “the size-dependent diffusional and gravitational losses for the inlet line were corrected by using the empirical functions”

(32) Page 23379, line 27: Change “was” to “were”

- (33) Page 23380, line 1: “according to the methods of ”
- (34) Page 23380, line 6: submicron particle mass-size distributions”
- (35) Page 23380, line 10: “(e.g., NO, NO_x, CO, O₃ and SO₂)”
- (36) Page 23380, lines 12-13: “were given by Tang et al. (2012) and Wang et al. (2014b).”
- (37) Page 23380, lines 15-17: “was used to obtain meteorological parameters (e.g., relative humidity, air temperature, wind speed and direction). The time base for all data in this study was Beijing zone time (=UTC+8)”
- (38) Page 23380, lines 20-26: “Figure 1 shows the time series of ...during the period. The average mass concentrations of ..., respectively, indicative of the high level of aerosol pollution. The average mass ratios of..., respectively. As we can see in Fig. 1b, the mass ratio of PM₁/PM_{2.5} is higher than that of PM_{2.5}/PM₁₀ before 28 January, indicating that PM₁ dominated the total mass of aerosol particles.”
- (39) Page 23381, line 1: Please move the “lines 20-28” before the “Figure S1 in the Supplement displays...”.
- (40) Page 23381, line 1: Figure S1 in the Supplement did not give the marked scale of the wind speed.
- (41) Page 23381, lines 2-5: “During this period, the average wind speed is 2.5 ms⁻¹. Figure S2 shows an overview of wind rose of the local wind and the wind is mainly in the southerly and northerly quadrant, which can bring relatively dirty or clean air masses,72h backward trajectories of air parcels...”
- (42) Page 23381, lines 8-9: “Beijing often suffers more polluted atmosphere than that in the northern area due to more dense cities and population. The clusters of 1 to 5 are from northern direction, with...”
- (43) Page 23381, line 10: Change “Also” to “Furthermore”
- (44) Page 23381, line 12: Change “local direction” to “local directions”, change the “most” to “highest”
- (45) Page 23381, line 16: Change “different color” to “different colors”
- (46) Page 23381, line 17: Change “at 1000 m to 1500 m” to “between 1000 m to 1500 m”

- (47) Page 23381, lines 18-19: “the lapse rate of temperature is nearly $-6^{\circ}\text{C}/100\text{m}$,”
- (48) Page 23381, line 21: “and $620.8 \mu\text{g}/\text{m}^3$, respectively.”
- (49) Page 23381, line 24: “showed opposite pattern with time...”
- (50) Page 23381, line 25: “compared with $\text{PM}_{2.5}$ ”
- (51) Page 23381, lines 26-28: “compared with PM_{10} with increasing aerosol pollution. It is worth noting that the increase of $\text{PM}_{1-2.5}$ was greatest during the period from 28 to 29 January, as showed in Fig. 1a.”
- (52) Page 23382, line 1: “3.2 Aerosol optical properties”
- (53) Page 23382, lines 3-7: “measured by the nephelometer. And the aerosol ...can be calculated from the scattering coefficients, which have rarely been reported in Beijing using in-situ measurements. The light scattering coefficients of aerosols show ...as mass concentrations of PM ...”
- (54) Page 23382, lines 8-18: “during this haze episode, and the average scattering coefficients....After converting the aerosol scattering coefficients at 550 nm to that of 525 nm, the average σ_{sp} at 525 nm are 3.2 times greater than the yearly average values at another urban site in Beijing, reported by He et al. (2009)...., respectively, as presented in Fig. 4b. During...three wavelengths are highly correlated. Both...increase gradually from 24 to 29 January and decrease sharply to lower levels, which are consistent with the variations of aerosol mass concentrations.”
- (55) Page 23382, lines 21-22: “by particles. It is related to particle size distributions, and can be calculated as following:”
- (56) Page 23382, line 25: Change “small sized particles” to “small size particles”
- (57) Page 23383, line 2: Change “can be” to “is”
- (58) Page 23383, line 5: Change “as follows;” to “as following:”
- (59) Page 23383, line 7: “and 0.94 ± 0.3 , respectively. The average...”
- (60) Page 23383, lines 8-10: “Which is smaller than that of 1.46 in Guangzhou (Garland et al., 2008) and 1.7 in Spain reported by Titos et al. (2012), which indicates a more dominant coarse mode particles compared with the other locations.”
- (61) Page 23383, lines 17-20: “between -1 of completely backscattered light to +1 for

completely forward scattered light. Because there is no measurements can be directly obtained the values of g , a fit equation applied by Andrews et al. (2006) was used as in Eq. (4).”

(62) Page 23383, line 23: “ 0.54 ± 0.05 , respectively.”

(63) Page 23384, lines 1-5: “..shows higher values, which g_λ shows lower ones, as showed in Fig. 4. However, the opposite feature occurs when the haze developed. Especially during the highest pollution periods (from 28 to 30 January), higher values of g_λ and lower values of b_λ and $\overset{\circ}{A}_{SP}$ appear, ...”

(64) Page 23384, lines 5-6: “which indicates an increasing fraction of relative coarse aerosol, consistent with the variation pattern of $PM_1/PM_{2.5}$ showed in Fig. 1b.”

⇒ Please give a short interpretation about this conclusion.

(65) Page 23384, lines 8-14: “The particle number-size distribution from 25 to 31 January is shown in Fig. 5. The particle number concentration peaks at a diameter of around 100 nm. These particles are mainly from direct emissions of vehicles,...(Shi et al., 2001). The time series of nucleation mode...concentrations are presented in Fig. 6. The Aitken mode particles shows the highest number concentrations during the period...”

(66) Page 23384, lines 16-27: “The lowest particle number concentration is in coarse mode....The Nucleation mode and accumulation mode also show high number concentrations,, respectively. Compared with that at another urban site in Beijing, ...and accumulation mode during this haze episode are 170 times, 10 times and 120 times, respectively (Hu et al., 2009). The nucleation and Aitken mode particles show a significant increase at mid-day on 28 January, while the accumulation mode is not significant. This may be ascribed to the emissions from vehicle and cooking nearby our sampling site. It is worth nothing that the concentration of coarse mode particle is highest on 29 January, ...”

(67) Page 23385, lines 1-2: “and Aitken mode particles decrease on 12:00 LT of 30 January, as shown in Fig. 6.”

(68) Page 23385, line 4: “The time series of chemical compositions, mass fractions,

O:C ratio and m/z 44 of NR-PM₁ are presented...”, please also give the explanations of O:C ratio and m/z 44.

(69) Page 23385, lines 7-10: “and $5.5 \pm 4.2 \mu\text{g}/\text{m}^3$, respectively. The organic component is dominant in NR-PM₁,Sulfate and nitrate species concentrations are also very high during the heavy haze event.”

(70) Page 23385, lines 11-15: “...size-resolved chemical compositions of different mode particles as a function of time. Figure 8 shows the temporal variations of the size distributions of the organic...and chloride (e). The organic and chloride containing particles display a slightly broader distribution than the other three species. All the aerosol components mainly....”

(71) Page 23385, lines 19-21: “Based on the research by Zhang et al. (2004) in Pittsburgh PA, USA, an average aerosol bulk density of 1.5 is assumed in this study. For a first approximation,to 470 nm in physical diameter of spherical particles.”

(72) Page 23385, line 24: Change “will lead to high light scattering” to “will lead to strong light scattering”

(73) Page 23385, lines 26-28: “These five aerosol components all show high28 January to noon of 29 January, corresponding....light scattering of the whole pollution period.”

(74) Page 23386, lines 2-3: “The particle number concentrations show a burst...”

(75) Page 23386, line 5: “formation event, which was accompanied by advection of local emissions.”

(76) Page 23386, line 7: “on 28 January is observed by AMS as shown in Fig. S4. The...”

(77) Page 23386, lines 9-11: “This may be due to the accumulation of air pollutants under the stagnated boundary layer. As we can see in Fig. S1, the meteorological parameters are characterized...”

(78) Page 23386, lines 13-15: “the dilution causes the aerosol concentrations decreasing in the afternoon. The concentrations of sulfate, ammonium and nitrate show an increasing trend from 18:00 LT. The major reasons are: (1) Increasing RH...”

(79) Page 23386, lines 19-20: “All of the above aspects result in the mass

concentrations of sulfate and ammonium have a distinct the growth of particles with diameters between 100 nm and 500 nm on 28 January.”

(80) Page 23386, line 23: “Figure 10 shows the variations of signal...”

(81) Page 23386, lines 25-27: “...are presented as well. The highest....aerosol concentration appears nearly between 20 to 35 $\mu\text{g}/\text{m}^3$, ...The signal of m/z 44 shows an increasing trend...”

(82) Page 23387, lines 1-4: “component mainly exists at RH below 40%, which is indicative of ...in urban Beijing. It is notable that the higher levels of the organic component occurs under high RH conditions, of which aerosol water uptake ability is enhanced and the more highly hydrated particles are able to...”

(83) Page 23387, line 6: Change “increased” to “increases”

(84) Page 23387, line 8: “also show that aqueous-phase processes are responsible for the production of ...”

(85) Page 23388, line 15: Delete the “can be”.

(86) Page 23388, line 18: “AMS can only provide us with mass concentrations...”

(87) Page 23388, lines 27-28: “In IMPROVE algorithm, ..., while the contribution of organic aerosol didn’t take into account.”

(88) Page 23389, line 10: “as shown in Fig. 13...”

(89) Page 23389, lines 19-22: “Yao et al. (2010) showed that the organic components were contributed greatly to the light extinction (about 45% contribution) by using AMS data during winter in Shenzhen, PRC. Waston (2002) also found the organic aerosol dominated light extinction in some cities, with fractions of 9-50% in eastern USA.”

(90) Page 23390, lines 2-7: “Based on in-situ measurements, the physical and chemical properties of...were characterized during a severe haze episode in Beijing from 24 to 31 January, 2013. The average... $265.2 \pm 157.1 \mu\text{g}/\text{m}^3$, respectively, and an increasing.....during the most heavy pollution period. The average scattering coefficient at 550 nm was....”

(91) Page 23390, line 14: “contributed to the growth of particle during the most...”

(92) Page 23390, lines 16-17: “High emissions of background pollutant combined

with....., which lead to enhance water uptake ability of summicron...”

(93) Page 23390, lines 23-24: “...also play an important role in visibility degradation during the winter haze episode in Beijing.”

(94) Page 23398: “**Table 1.** The statistics of aerosol optical properties during observation period.”, change the “Standard derivation” to “Standard deviation”

(95) Page 23399: “**Table 2.** The statistics of particle number concentration during observation period.”, change the “Standard derivation” to “Standard deviation”

(96) Page 23401: “**Figure 2.** The three days backward trajectories of air parcels during the observation period. The colors of air trajectories represent the height during transport.”

(97) Page 23408: The Figure 9 is not analyzed or discussed in the manuscript, so delete the Figure.