

RESPONDS TO COMMENTS FROM REFEREE #1

First of all, we appreciate your very positive evaluation of our work. The responses of your specific comments/questions are outlined in detail below.

Specific comments:

(1) The analysis of long-term analysis of NPF and its frequency is incomplete without calculating and discussing the changes in the condensation (and coagulation) sink during this period — I highly recommend including this analysis. Furthermore, the authors could also calculate H₂SO₄ proxy (Dada et al., 2020) to estimate the changes in the NPF precursors over the study period.

Response: Thank you for your constructive suggestions and providing important reference. As you said, the condensation (and coagulation) sink and the precursors were very important for analyzing NPF and its frequency, which should be included in this study.

Referred to the methods presented in Dal Maso et al. (2005), NPF events were identified for a day. Number concentration sharply increased in the nucleation mode size range (13-25 nm) and prevailed for at least an hour. Additionally, the particle size was required to increase during the next few hours. The parameters describing NPF events such as formation and growth rates (J_D , GR and hereafter), condensation and coagulation sink (CS , $CoagS$ and hereafter) were calculated in this study. GR can be calculated with the time evolution of geometric mean diameter (GMD) of the nucleation mode obtained by parameterizing PNSD, and it can be expressed as

$$GR = \frac{dGMD}{dt} \quad (1)$$

The formation rates (J_D) can be calculated by the below equation:

$$J_D = \frac{dN_{nuc}}{dt} + F_{coag} \quad (2)$$

where the first term in the right hand side (dN_{nuc}/dt) represents the observed change of in number concentration of newly formed particles (Zhao et al., 2021). The second term is the loss of newly formed particles induced by coagulation scavenging, and can be obtained with the below equation:

$$F_{coag} = CoagS_{nuc} N_{nuc} \quad (3)$$

Coagulation sink of nucleation mode particles ($CoagS_{nuc}$) is defined as

$$CoagS(D_p) = \int K(D'_p, D_p) n(D'_p) dD'_p \quad (4)$$

where $K(D'_p, D_p)$ is the coagulation coefficient of particles with sizes of D_p and D'_p , calculated by the method of Fuchs (1964). The reference size (D_p) is assumed to be

the *GMD* of the nucleation mode. An average $CoagS_{nuc}N_{nuc}$ over each formation period was taken during the campaign.

The condensation sink (CS) can be expressed as

$$CS = 2\pi D \sum \beta_m(D_{p,i}) D_{p,i} N_i \quad (5)$$

where $D_{p,i}$ and N_i are particle diameter and the corresponding number concentration in size class i . D is the diffusion coefficient of the condensing vapor, usually assumed to be sulfuric acid. β_m represents a transition-regime correction (Kulmala et al., 2012),

$$\beta_m = \frac{1 + Kn}{1 + 1.677Kn + 1.333Kn^2} \quad (6)$$

defined as a function of the Knudsen number, $Kn = 2\lambda/D_{p,i}$. Furthermore, based on the method presented in Dada et al. (2020), H_2SO_4 proxy was calculated to estimate the changes in the NPF precursors over the study period, and the equation was given as follows.

$$[H_2SO_4] = -\frac{CS}{2 \cdot (9.9 \times 10^{-9})} + \left[\left(\frac{CS}{2 \cdot (9.9 \times 10^{-9})} \right)^2 + \frac{[SO_2]}{(9.9 \times 10^{-9})} (1.6 \times 10^{-9} \cdot GlobRad) \right]^{1/2} \quad (7)$$

where CS was calculated by Equation (6). SO_2 concentrations are measured by the ultraviolet fluorescence method, and Global radiation (GlobRad) was measured by an SMP3 pyranometer (Kipp and Zonen, the Netherlands) during the campaign. In addition, the peak sizes of PNSDs are determined as mode diameters.

To better analyze long-term trend of NPF event and the relevant parameters during 2012-2019, Figure 11 illustrates the inter-annual statistics of the trends of NPF frequency, mode diameter, and formation and growth rates. Furthermore, condensation and coagulation sink (CS , $CoagS$) and H_2SO_4 proxy were also calculated over the study period. Similar with the opposite N_{13-25} trend between the two contrasting periods (Figures 8-9), the occurrence frequency of NPF events decreased from $\sim 30\%$ to less than 5% until 2016 and then increased to more than 30% in 2019. The particle has been becoming much finer since 2015 due to more frequent NPF events (Figure 11B). The temporal variations of PNCs in nucleation mode (dN_{nuc}/dt , Figure 11C) and coagulation scavenging effect (F_{coag} , Figure 11D) followed similar inter-annual variations of NPF frequency. The contribution of coagulation loss flux F_{coag} to total observed rate was on average 37% , which was close to the average ratio of coagulation loss to formation rate in urban Beijing, 0.41 (Yue et al., 2010), suggesting that coagulation loss was the same important as dN_{nuc}/dt . The formation rate (J_D) ranged from 0.2 to $16.2 \text{ cm}^{-3} \text{ s}^{-1}$ in urban Lanzhou, which was lower than the observations at some urban sites, such as in Beijing, $3.3\text{-}81.4 \text{ cm}^{-3} \text{ s}^{-1}$ (Wu et al.,

2007), St. Louis, with the mean value of $17.0 \text{ cm}^{-3} \text{ s}^{-1}$ (Qian et al., 2007), but much higher than that in regional nucleation episodes $0.01\text{-}10 \text{ cm}^{-3} \text{ s}^{-1}$ at the most other sites (Kulmala et al., 2004).

Compared with J_D , GR varied less in inter-annual scale, and ranged from 0.5 to 14.9 nm h^{-1} , slightly higher than that in urban Beijing, $0.3\text{-}11.2 \text{ nm h}^{-1}$ (Wu et al., 2007), and also within the range of typical particle growth rate $1\text{-}20 \text{ nm h}^{-1}$ in mid-latitudes (Kulmala et al., 2004). The inter-annual variation of condensation sink (CS) was consistent with that of NPF frequent and formation rate with the range between 7.3×10^{-4} and $5.1 \times 10^{-2} \text{ s}^{-1}$ with mean value of $1.4 \times 10^{-2} \text{ s}^{-1}$ (Figure 11F), which was comparable with the calculated value of 0.02 s^{-1} during NPF events in the North China Plain (Shen et al., 2011). Based on the method presented in Dada et al. (2020), we also calculated H_2SO_4 proxy to estimate the changes in the NPF precursors over the study period (Figure 11G). The H_2SO_4 proxy varied from 3.3×10^7 to $6.0 \times 10^8 \text{ cm}^{-3}$ with average concentration of $2.5 \times 10^8 \text{ cm}^{-3}$ over the study period, which was slightly higher than that in urban Beijing (Dada et al., 2020) due to more coal combustion and basin terrain in urban Lanzhou. The used methods and the corresponding analyses and discussion will be included in the revised version of our manuscript.

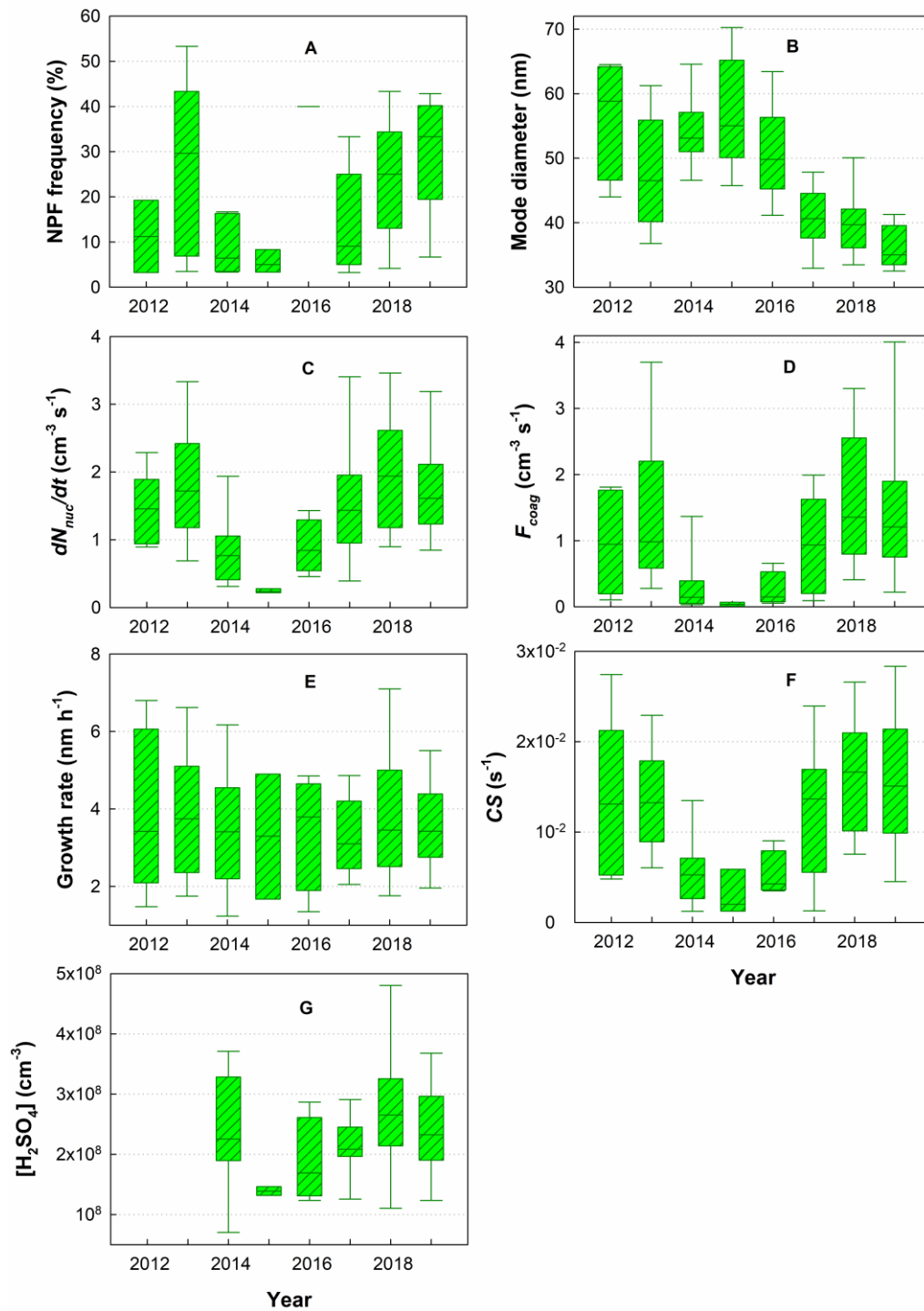


Figure 11: Inter-annual statistics of the trends of NPF frequency, mode diameter, formation (dN_{nuc}/dt , F_{coag}) and growth rates, CS and H_2SO_4 proxy during the campaign. The lines inside the box denotes the median slope, the two whiskers and the top and bottom of the box denote the 5th, 95th, 75th and 25th percentiles.

(2) The authors separate the study into two contrasting periods (before and after Jan 2016). However, based on the timeseries in Figure 2, it seems that 2013 is an unusually polluted year even for 2012–2015 period. I think presenting the average particle size distribution surface plots for each year (by season) can be instructive for highlighting the overall similarities and differences of each year of the study period (perhaps in the SI).

Response: Thank you for catching that. As you said, the average particle number size distribution (PNSD) surface plots in four seasons for each year during the campaign are presented in Figures S1-S4 to highlight the overall similarities and differences of each year during the study period. The mode diameter of PNSD shifts to smaller particle size in four seasons from 2012 to 2019. Particle number in Aitken and accumulation modes declined largely in autumn and winter during the study periods maybe due to the even strictest emission control policies in recent years. However, in spring and summer, the nucleation mode particle number increased significantly after 2016, which can be partly modulated by NPF events. Figures S1-S4 and the above corresponding discussions will be added to the revised version of our manuscript.

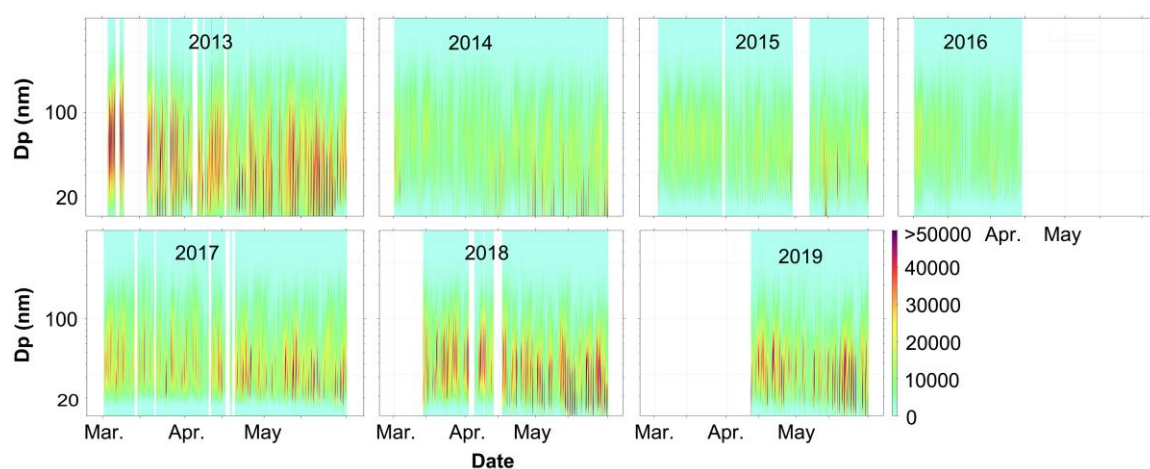


Figure S1: Evolutions of hourly average particle number size distributions (PNSD) in spring for each year during the campaign. The white gaps in the subplots represent missing data due to failures or routine maintenance of the instruments.

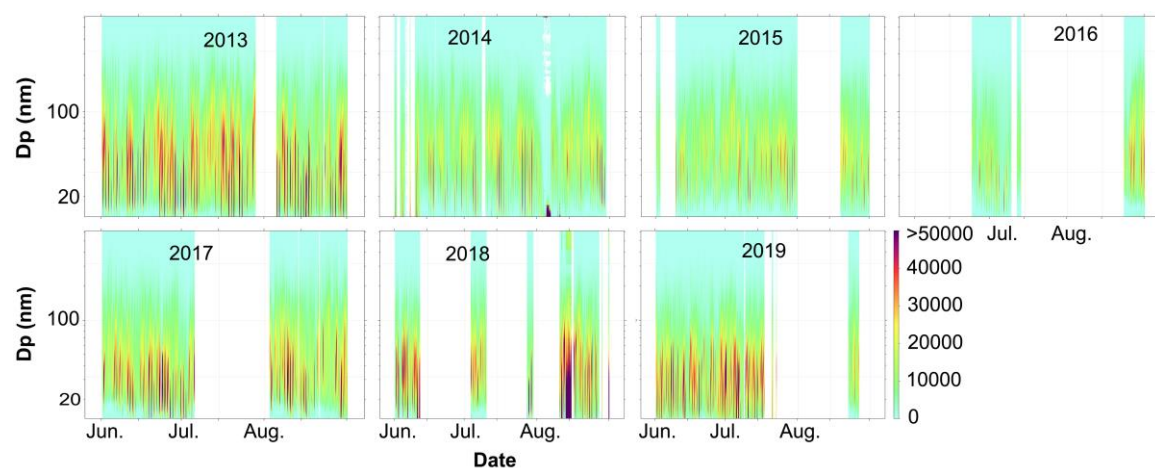


Figure S2: Evolutions of hourly average particle number size distributions (PNSD) in summer for each year during the campaign. The white gaps in the subplots represent missing data due to failures or routine maintenance of the instruments.

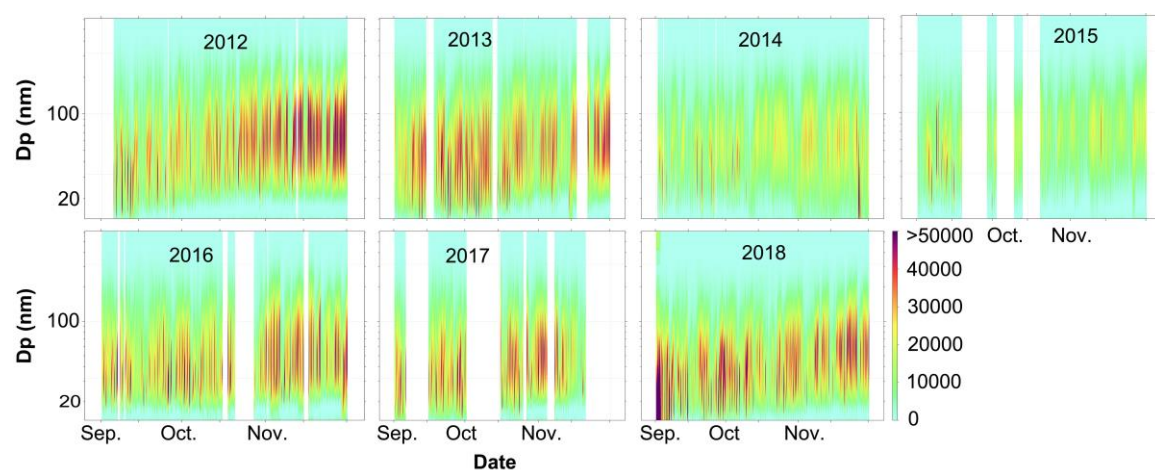


Figure S3: Evolutions of hourly average particle number size distributions (PNSD) in autumn for each year during the campaign. The white gaps in the subplots represent missing data due to failures or routine maintenance of the instruments.

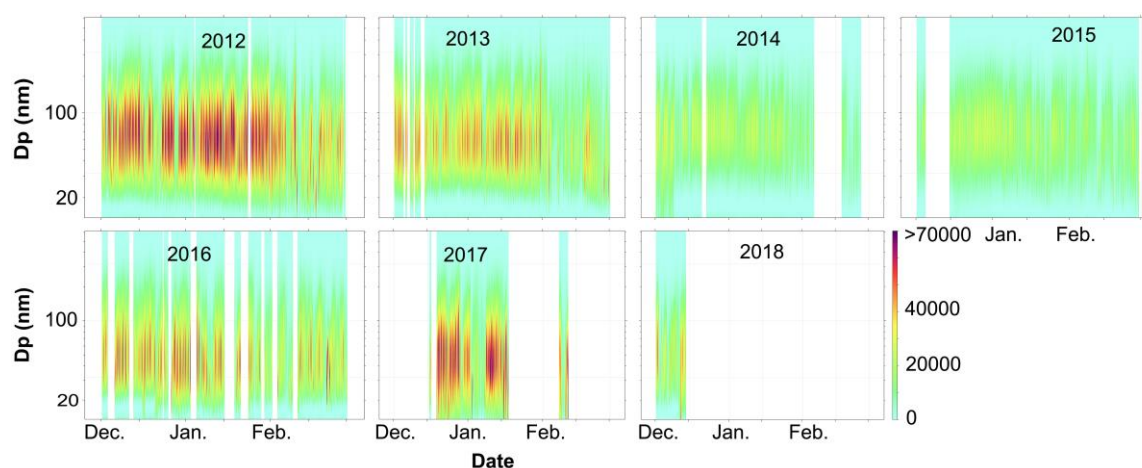


Figure S4: Evolutions of hourly average particle number size distributions (PNSD) in winter for each year during the campaign. The white gaps in the subplots represent missing data due to failures or routine maintenance of the instruments.

(3) Many of the graphics in the manuscript are well made and explain the central themes of the study well (e.g., I think Figure 4 is excellent). However, I think the readability of manuscript can be improved if some of the graphics that are supplementary to the analysis are removed or moved to the SI (e.g., Figure 3). Furthermore, some more details should be provided in the captions of the figures as they should be interpretable independent of the text (e.g., Figure 9 was not clear to me).

Response: Thank you for your suggestions. To improve the readability of manuscript, Figures 3, 5, 11, 15 and 16 that are supplementary to the analysis will be moved to the SI. Furthermore, some more details will be provided in the captions of the all figures.

(4) Page 9, Line 4 (“The nucleation mode particles also can grow...”). I do not think this sentence is correct or required for this discussion. The timescales for such a transport (from ground to the sampling inlet) is likely to be much shorter than for the particles to grow from nucleation to Aitken mode.

Response: Thank you for your reminder. The sentence in Line 4 of Page 9 will be deleted in the revised version of our manuscript.

(5) Page 17, Line 15 (“NPF events predominantly occurred under dry and sunny weather conditions”). This should be discussed with more nuance based on more recent literature. For example, according to a relatively recent review paper on NPF, “The observed factors that favor the occurrence of regional NPF include a high intensity of solar radiation, low RH, high gas-phase sulfuric acid concentration, and low pre-existing particle loading, i.e. low CS and CoagS” (Kerminen et al., 2018).

Response: Thank you for your good suggestions. According to a relatively recent review on regional NPF in different environments of the global troposphere, the observed factors that favor the occurrence of regional NPF include a high intensity of solar radiation, low RH, high gas-phase sulfuric acid concentration, and low pre-existing aerosol loading (Kerminen et al., 2018). The possible reasons for the apparently close connection between the ambient RH and occurrence of NPF have been proposed, including the typically negative feedback of high RH on the solar radiation intensity, photochemical reactions and atmospheric lifetime of aerosol precursor vapors. The effect of the ambient temperature (T) on NPF shows very different responses between different studies, which is probably related to the simultaneous presence of several temperature-dependent processes that may either enhance or suppress NPF. Therefore, the meteorological parameters affect NPF process by modulating the condensation and coagulation sink. We will add the above discussion to the revised manuscript.

(6) For figures in manuscript or SI: (i) Include units of all parameters (where applicable); (ii) Avoid using captions such as “Same as Figure X, but for...”; (iii) Use continuous colorbars when using surface plots (Fig. 17 and Fig. S7).

Response: Thank you for your good suggestions. The all figures and the corresponding captions in new manuscript and SI will be revised according to your three suggestions.

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RESPONDS TO COMMENTS FROM REFEREE #2

First of all, we appreciate your very positive evaluation of our work. The responses of your specific comments/questions are outlined in detail below.

Specific comments:

(1) Climate and environmental effects of aerosol particles largely depended on their optical properties, and thus the variations in optical parameters during the measurement campaign should be explained or referred to some the published important works in Introduction. AOD and Alpha, two important optical parameters of aerosol particles, were analyzed during the campaign. However, the statement of relevant instrument should be given in the section of “Data and methods” of the revised manuscript.

Response: Thank you for your suggestion. As you said, aerosols can affect the local climate and environment by light absorption or scattering, which significantly depended on chemical composition and size distribution of aerosol particles. Liu et al. (2020) indicated that coating plays an important role in light absorption. The amplification of black carbon absorption by the coating increased from 1.21 to 1.75 with increasing aerodynamic diameter (D_{ae}) due to the thicker coating of BC-containing particles with a larger D_{ae}. Their study highlights the strong dependence of the microphysical and optical properties of BC on size. The more recent study of Zhao et al. (2021) found that interdecadal AOD was negative trend from 2009 to 2018, which may be related to the variation in particle size distribution. AOD and Alpha were measured continuously by CE-318. The above discussions and the previous studies will be added to the revised manuscript.

(2) The limitations on used data such as only a sampling site and used analyzing method should be given, and future works should be preliminarily introduced by 1-2 sentences in the last paragraph of the conclusions.

Response: Thank you for your good suggestion. In this study, the measurement campaign was conducted at a Chinese cities in west China, but the similar PNCs trends and influencing factors should be expected in other Chinese cities. In future work, we will established the PNSD observation network in some megacities to better evaluate the response of PNCs to emission mitigation policies in China. The above explanation will be added to the revised manuscript.

(3) All abbreviations should be defined when they firstly appear throughout the manuscript, such as NPF, particle number in different size bins (N13-25, N25-100, N100-800).

Response: Thank you for catching that. All abbreviations will be defined when they firstly appear throughout the revised manuscript.

(4) The sampling site should can represent urban background. Please explain that in the section of “Data and methods”.

Response: Thank you for your question and suggestion. The sampling site can represent urban background, which will be explained in the “Data and methods” of the revised manuscript.

(5) The formation and evolution of air pollution may be more complex inside the basin as compared to that at the plain due to terrain and PBL impacts. Therefore, difference of aerosol pollution between inside the basin and the plain and the climate and environmental effects should be introduced in detail in the section of Introduction.

Response: Thank you for your good suggestion. The atmospheric horizontal and vertical dispersion conditions inside the basin were poor due to weak winds and strong multi-layer temperature inversion induced by basin terrain (Pandolfi et al., 2014). Therefore, the air pollutants were easily trapped inside the basin and hard to disperse to the upper air. Furthermore, basin aerosol pollution was largely controlled by vertical than horizontal dispersion as compared to the plain (Zhao et al., 2019). We will add the above explanations to the section of Introduction in the revised version of our manuscript.

(6) Only some novel findings should be clearly listed in the conclusions, and thus that should be revised to provide some key conclusions.

Response: Thank you for your good suggestions. The conclusions will be revised and some novel findings will be clearly listed in the revised version of our manuscript.

(7) All figures in the manuscript should be revised and largely improved for seeing more clearly by readers.

Response: Thank you for your constructive suggestion. As you said, all figures are not easily seen by the readers and they will be revised in the new manuscript.

References

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