Response to reviewers

Thanks for your great efforts and valuable comments, which helps to improve our manuscript. We have addressed the reviewers' comments on a point-to-point basis as below for consideration. Referee comments are in black. Author responses are in red. All the line numbers mentioned following are refer to the revised manuscript with no changes marked.

- **RC2**: 'Comment on acp-2022-837', Anonymous Referee #2, 06 Feb 2023

  This manuscript entitled 'Characterization of dust-related new particle formation events based on long-term measurement in North China Plain', describes the observation of new particle formation events following dust episodes observed in Beijing between 2017 and 2021, with a focus on one severe episode on March 15-16, 2021. Although the topic of the paper and the potential results regarding the interaction of dust with available precursors resulting in NPF could be an important addition to literature, the authors' claims that are not supported by scientific evidence prevent me from recommending this paper for publication in ACP.

  General comments:

1. The authors evaluate the contribution of anthropogenic emissions to the nucleation processes.
   The authors calculated the anthropogenic emissions as the difference in the size distribution, formation rates and growth rates during other NPF days and NPF following dust days. This approach is not justified, as it ignores available sinks, meteorology and atmospheric reactions.

   Reply: The authors agree with the reviewer's comment that quantifying the contribution of anthropogenic emissions to NPF events based on the comparison of dust related NPF and non-dust NPF days was not robust and justified, due to the lack of direct measurement of precursors and theoretical calculations. However, clear differences between dust related and non-dust related NPF events were found based on the long-term PNSD measurement, providing valuable information about how the dust storm process modified the atmosphere conditions when NPF event occurred. According to the reviewer's comments, we removed the conclusions about the quantifying of the influence of anthropogenic emissions on NPF and focused on how the dust process affecting NPF and highlight the intrinsic reasons.

2. Sections 2.3-2.5 discuss a case study in March 15-16, 2021. While the results are interesting, the conclusion cannot be extrapolated to other events as the case study described is one of a kind in the four years, as shown in Table 2. A comparison between NPF events following different types of dust episodes events could be helpful. In particular, a comparison between the episodes on March 15 and 28, with

including interpretation of NPF and its related properties could give insight on the effect of different types of dust.

Reply: Thanks for your comments, we added the discussion of NPF events followed different types of dust days. We found the dust related NPF events are highly dependent on the strength and the end time of dust process. NPF event usually occurred around 8:00-12:00 when solar radiation increased, so if the dust processes prevailed during this time, it will prevent NPF occurrence. For some cases of blowing and floating dust days overlaid by the anthropogenic aerosols, for example, March 18, 2019, April 5, 2019 and May 15, 2019, the *CS* remains high during the dust (above 0.02 s^{-1}), which prevented NPF event. Under some cases (April 15-16, 2021, Fig. S1), the whole dust process was identified as blowing and floating dust episode depending on the strength how air masses containing dust particles influenced Beijing. NPF event can be observed when the whole dust process finished.

Blowing dust and floating dust was observed on May 6-7 and May 8, respectively, and both followed by the occurrence of NPF event, as shown in Fig. S2. It was also found the condensation sink (*CS*) before NPF started was approximately 0.0025-0.003 s^{-1} for both NPF events, indicating the extremely weak scavenging process by the pre-existing particles. The *CS* level were also similar during the NPF event indicating the concentration level of precursors and condensing vapor participating nucleation and growth were comparable for these two cases. The values of formation rate (J_3) and growth rate (*GR*) of NPF event on May 7th and 8th were quite close, with J_3 of 4.5 and 4.8 $cm^{-3}s^{-1}$, *GR* of 2.3 and 2.5 nm h^{-1}, respectively.

The discussions are supplemented in the manuscript and the figures are given in the supplementary materials.

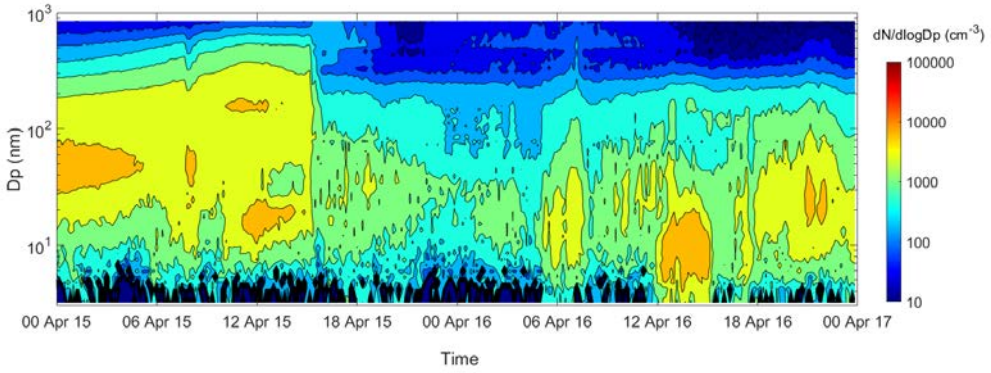

Fig. S1, NPF event occurred following blowing dust (April 15) and floating dust (April 16) in 2021

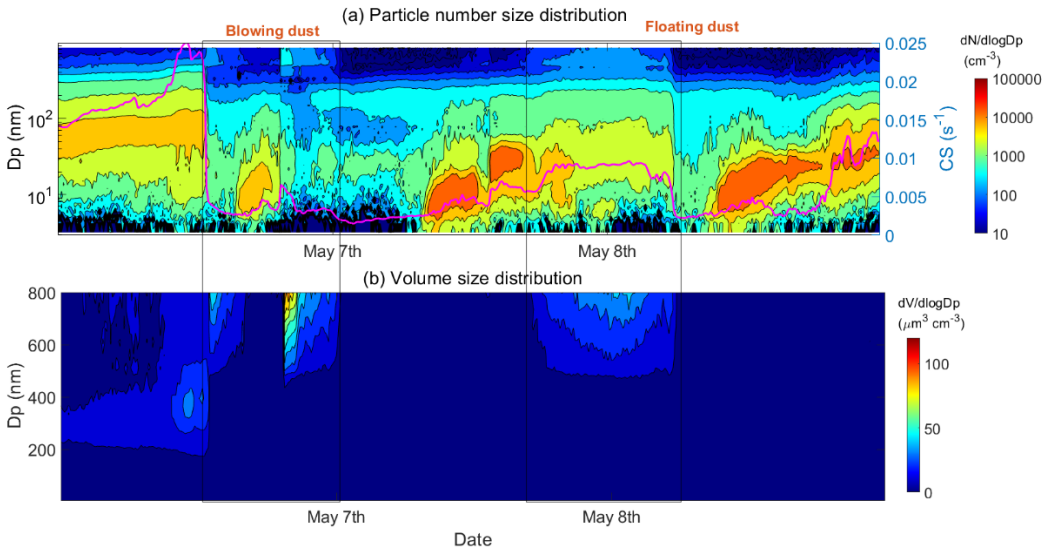

Fig. S2, NPF event occurred following blowing dust (May 6 and 7) and floating dust (May 8) in 2021. The particle number (a) and volume (b) size distribution are given and *CS* is given by pink line.

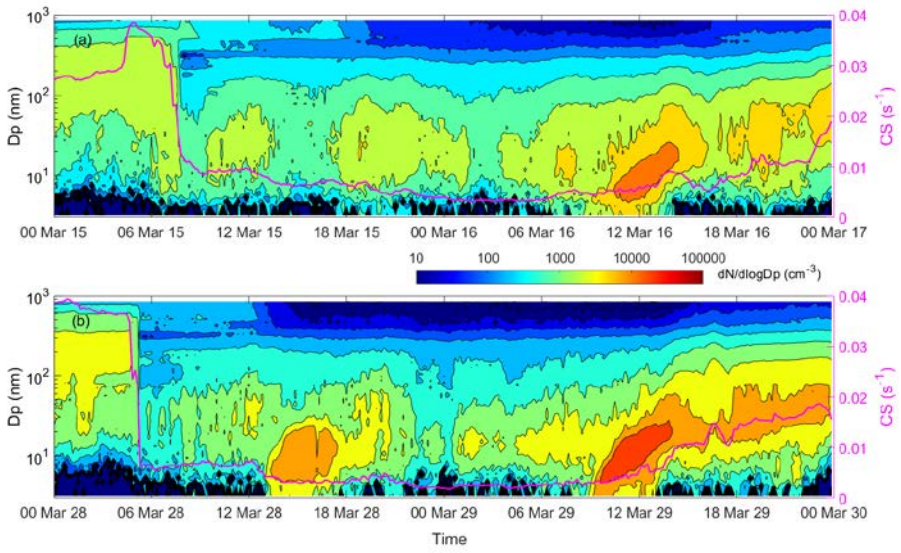

Fig. S3 The NPF event occurred after dust storm on March 15 (a) and blowing dust on March 28, 2021 (b), respectively. The pink line in panel indicates the condensation sink.

3. The authors report that an NPF event occurs after approximately 80% of dust episode, what was the limiting factor for the remaining 20%? Was the airmass direction different, the wind speed lower etc? Identifying the causes that inhibited the NPF during those 20% could improve the quality of the results.

Reply: Thanks for the constructive suggestion. We checked through all the dust days with NPF and without NPF days. It was found the air masses changed from northwest with the dust sources to other directions with anthropogenic emissions dominated. As given in Fig. S4-S5, floating dust occurred on May 15 2019 and April 1 2021, without

NPF event when dust diminished. It showed the air mass originated from northwest during dust period on 00 UTC and switched to southwest since 06 UTC on May 15 and southeast since 06 UTC on April 1, respectively. The change of air masses resulted in the polluted dust case, which is the mixture of anthropogenic aerosols and dust and NPF event was prohibited due to the high level of condensation sink. Even the nucleation process was observed on April 1, the growth process was interrupted by the elevated background aerosol concentration, indicated by the increasing *CS*.

The 72 hours back trajectories arriving at CAMS station at four times a day (00, 06, 12, 18 UTC) on May 15 2019 and April 1 2021, respectively, with the terminal height of 500 m above ground level, derived by the TrajStat software, combined with HYSPLIT 4 model (Draxler and Hess, 1998; Wang et al., 2009). Trajstat, is a geographic information system-based software, which can view, query, and cluster the air mass trajectories and also conduct the potential source contribution analysis (Wang et al., 2009).

The above information has been added in the manuscript and supplementary materials.

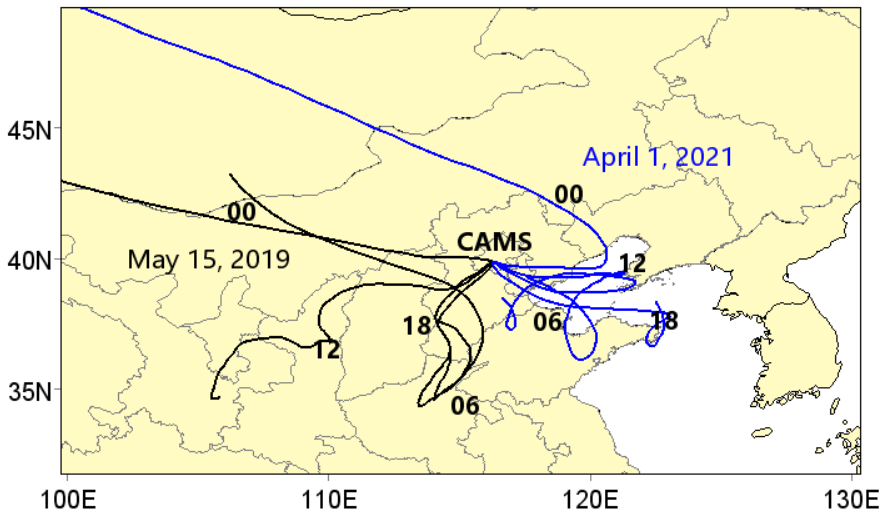

Fig. S4. The 72 hours back trajectories arriving at CAMS station at four times a day (00, 06, 12, 18 UTC) on May 15th 2019 (black lines) and April 1st 2021 (blue lines) with the terminal height of 500 m above ground level, derived by the TrajStat software, combined with HYSPLIT 4 model

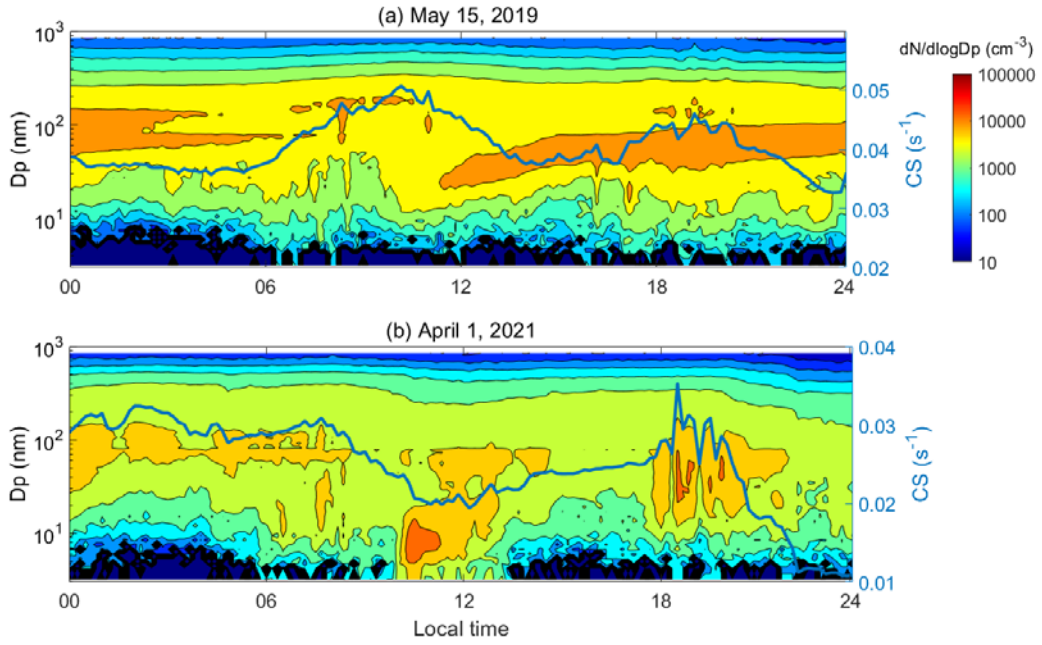

Fig. S5. The cases of dust episode (May 15, 2019 and April 1, 2021) without typical NPF followed.

Draxler, R.R., Hess, G.D., 1998. An overview of the HYSPLIT_4 modeling system for trajectories, dispersion, and deposition. Aust. Meteorol. Mag. 47 (4), 295–308.

Wang, Y.Q., Zhang, X.Y., Draxler, R.R., 2009. TrajStat: GIS-based software that uses various trajectory statistical analysis methods to identify potential sources from long-term air pollution measurement data. Environ. Model. Software 24 (8), 938–939. https://doi.org/10.1016/j.envsoft.2009.01.004.

4. In section 3.3, the authors speak about primary and secondary organics, how did the authors derive this separation? Were there source apportionment techniques involved? Or was it based on previous literature? More details on the technique and results are needed.

Reply: The primary and secondary organic aerosols are identified based on the AMS data, by applying PMF method. The details are supplemented in the section 2.2 instrumentation.

Positive matrix factorization (PMF) (Ulbrich et al., 2009) and a multilinear engine (ME-2) (Canonaco et al., 2013) modelling of high time resolution organic mass spectrometric data from HR-ToF-AMS have also been used to resolve organics into primary organic aerosols (POA) and oxygenated organic aerosols (OOA), which correspond to different sources and processes (Zhang et al., 2022).

Canonaco, F., Crippa, M., Slowik, J. G., Baltensperger, U. and Prévôt, A. S. H.: SoFi, an IGOR-based interface for the efficient use of the generalized multilinear engine (ME-2) for the source apportionment: ME-2 application to aerosol mass spectrometer data, Atmos. Meas. Tech., 6(12): 3649-3661, DOI: 10.5194/amt-6-3649-2013, 2013.

Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R. and Jimenez, J. L.: Interpretation of organic components from Positive Matrix Factorization of aerosol mass spectrometric data, Atmos. Chem. Phys., 9(9): 2891-2918, DOI: 10.5194/acp-9-2891-2009, 2009.

Zhang, Y., Zhang, X., Zhong, J., Sun, J., Shen, X., Zhang, Z., Xu, W., Wang, Y., Liang, L., Liu, Y., Hu, X., He, M., Pang, Y., Zhao, H., Ren, S. and Shi, Z.: On the fossil and non-fossil fuel sources of carbonaceous aerosol with radiocarbon and AMS-PMF methods during winter hazy days in a rural area of North China plain, Environ Res, 208: 112672, DOI: 10.1016/j.envres.2021.112672, 2022.

5. I suggest that the authors revise some of the interpretations in the paper which are either not consistent or not based on evidence from the observations. Examples:

Reply: we agree with the reviewer's comments that some conclusions in the manuscript were nor robust, and we have revised them based on the measurement data directly. Some sentences are not helpful to discussion, which are removed from the manuscript.

- Line 241: "When the dust particles arrived in Beijing at 8:00 LT on March 15, the volume mixing ratio of NO_2 and SO_2 decreased immediately, which was also influenced by the enhanced aerosol surface uptake process owing to elevated particle surface concentration" vs Line 247: "However, the concentrations of NO_2 and SO_2 were low during dust storms, indicating that anthropogenic emissions had less influence."

Reply: original L241: it has been revised to "When the dust particles arrived in Beijing at 8:00 LT on March 15, the volume mixing ratio of NO_2 and SO_2 decreased immediately due to the strong dilution by wind".

original L247: this sentence was removed.

- Line 284: During the dust storm period, transitional metal ions such as Fe and Mn can act as catalysts that favor sulfate formation via SO_2 oxidation (Usher et al 2003).

Reply: This sentence was removed.

- Line 262: The decrease in NO_2 and nitrate suggests that they probably shifted from fine to coarse particles during dust storms (Wang et al., 2013).

Reply: As we do not have direct chemical composition measurement, this sentence is removed.

- Line 269: This indicates different sinks for SO_2 and NO_2 during the dust storms. As discussed above, the uptake by mineral dust was a major sink for SO_2, whereas the uptake of NO_2 was minor, as the concentration remained low when the dust storm ended.

Reply: This paragraph has been revised to "The SO_2 volume mixing ratio increased quickly when the dust storm faded (22:00 LT on March 15), which was probably due to the weakened dilution process as the wind speed decreased from 5.8 m/s during dust episode to below 3.0 m/s at 20:00 LT on March 15. Another possibility was indicating that dust particles could be a major SO_2 sink (Usher et al., 2003). NO_2 also decreased when the dust storm started; however, it did not increase significantly when the dust storm ended, which differed from the variation in SO_2. However, heterogeneous reactions of mineral dust with SO_2, NO_2 can not be discussed further, due to the lack of direct measurement of chemical information of coarse mode particles."

6. The methods section lacks detailed explanation of most of the instrumentation and quality control.

- Were the aerosol measuring instruments cross checked for over-lapping sizes? Citations are missing.

Reply: The details of instrumentation, experimental setup and data quality control, as well as the citations, are supplemented in the text.

As the reviewer mentioned, there is an overlap size range of particle number size distributions (PNSDs) derived from TSMPS and APS, which is 500-850 nm. The resulting distributions of APS system were converted from aerodynamic to mobility diameters with assumed particle density of 2.5 g/cm^3 during the dust, as combined with TSMPS data. The PNSDs and calculated volume size distribution derived from TSMPS and APS, respectively, during March 15-17, 2021 was given in Fig. S6. The volume concentration was calculated based on PNSD, with the assumption of spherical shape. It showed the submicron particles (PM_1) was the dominant contributor to the particle number concentration (a), whereas the contribution to the volume can be ignored (b). It was found in the overlap size range between TSMPS and APS data (500-850 nm), the bias became smaller when particle size increased. At the last size bin of 850 nm, the number concentration derived from TSMPS was 50% higher than that from APS. The information of the overlap size range was supplemented in the text and the figure was given in the supplementary materials.

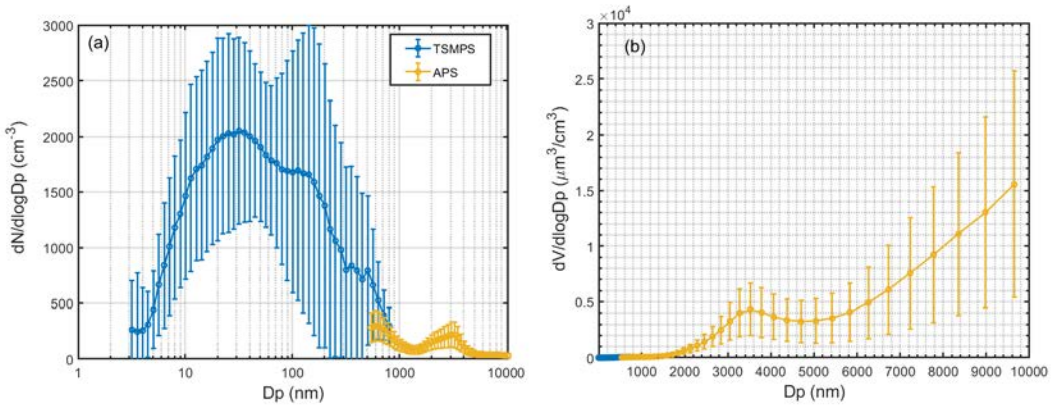

Fig. S6. The number size distribution (a) and volume size distribution (b) derived from TSMPS and APS.

The description of H-TDMA measurement and data inversion has been also added. The H-TDMA system is comprised of two DMAs, a CPC (Model 3772, TSI Inc., USA) and a humidifier system between the two DMAs. The first DMA selects the quasi-monodisperse particles at a diameter ($D_{p,dry}$ = 50, 100 nm) under the dry state with 30% RH (Maβling et al., 2003). Then, the size-selected particles pass through a humidity conditioner, which can be adjusted to the setting RH of 90%. The probability distribution function (PDF) of hygroscopic growth factor (HGF), HGF-PDF is inverted by the TDMAinv method developed by Gysel et al. (2009).

Maβling, A., Wiedensohler, A., Busch, B., Neusüß, C., Quinn, P., Bates, T., Covert, D.: Hygroscopic properties of different aerosol types over the Atlantic and Indian Oceans. Atmos. Chem. Phys. 3, 1377–1397, 2003.

Gysel, M., McFiggans, G.B., Coe, H.: Inversion of tandem differential mobility analyser (TDMA) measurements. J. Aero. Sci. 134–151, 2009.

- What are the detection limits and the time resolutions of the trace gas measuring instruments?

Reply: The TE 49C has a lower detection limit of 1 ppb and a precision of 1 ppb. The 42 CTL has a lower detection limit of 50 ppt and a precision of 0.4 ppb. The 43 CTL has a lower detection limit of 0.10 ppb and a precision of 1 ppb (Lin et al., 2009). Measurement signals of trace gases were recorded as 1 min averages (Lin et al., 2011), however, the hourly average data were used for discussion, in order to match with the PM mass concentration data. The above information and references have been added in the manuscript.

Lin, W., Xu, X., Ge, B. and Zhang, X.: Characteristics of gaseous pollutants at Gucheng, a rural site southwest of Beijing, Journal of Geophysical Research, 114, DOI: 10.1029/2008jd010339, 2009.

Lin, W., Xu, X., Ge, B. and Liu, X.: Gaseous pollutants in Beijing urban area during the heating period 2007–2008: variability, sources, meteorological, and chemical impacts, Atmospheric Chemistry and Physics, 11(15): 8157-8170, DOI: 10.5194/acp-11-8157-2011, 2011.

- The description of the HR-AMS-ToF is not sufficient, and citations are missing. For example what was the collection efficiency?

Reply: The description of HR-ToF-AMS with the citations has been supplemented in the manuscript. The chemical composition of non-refractory PM_1, including organic components, sulfate, nitrate, ammonium, and chloride, was derived using HR-ToF-AMS with a 5-min resolution (Drewnick et al., 2005). The calibrations of ionization efficiency were performed, using size-selected (300 nm) ammonium nitrate particles before and after the experiment. Default relative IE values were used for organics (1.4), nitrate (1.1), sulfate (1.2), ammonium (4.0), and chloride (1.3). The HR-ToF-AMS collection efficiency (CE) accounts for the incomplete detection of aerosol species owing to particle bounce at the vaporizer, and/or the partial transmission of particles by the lens (Canagaratna et al., 2007). In this study, a composition-dependent CE correction was used, following the methodology described by Middlebrook et al. (2012).

Canagaratna, M. R., Jayne, J. T., Jimenez, J. L., Allan, J. D., Alfarra, M. R., Zhang, Q., Onasch, T. B., Drewnick, F., Coe, H., Middlebrook, A., Delia, A., Williams, L. R., Trimborn, A. M., Northway, M. J., DeCarlo, P. F., Kolb, C. E., Davidovits, P., and Worsnop, D. R.: Chemical and microphysical characterization of ambient aerosols with the aerodyne aerosol mass spectrometer, Mass Spectrometry Reviews, 26, 185-222, 10.1002/mas.20115, 2007.

Drewnick, F., Hings, S. S., Decarlo, P., Jayne, J. T., Gonin, M., Fuhrer, K., Weimer, S., Jimenez, J. L., Demerjian, K. L., and Borrmann, S.: A New Time-of-Flight Aerosol Mass Spectrometer (TOF-AMS)—Instrument Description and First Field Deployment, Aerosol Science & Technology, 39, 637-658, 2005.

Middlebrook, A. M., Bahreini, R., Jimenez, J. L., and Canagaratna, M. R.: Evaluation of Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass Spectrometer using Field Data, Aerosol Science and Technology, 46, 258-271, 10.1080/02786826.2011.620041, 2012.

- Was there source apportionment performed?

Reply: The source apportionment has been conducted and the details are given in the supplementary materials.
The potential source contribution function (PSCF) analysis method has been widely applied to study the potential source regions of pollutants (Ashbaugh et al., 1985; Wang, et al., 2009). The PSCF values for each grid cell (0.5°*0.5°) in the selected domain were calculated by counting the number of trajectories those terminated within each grid cell, as follows:

$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}} \quad (1)$$

where n_{ij} is the number of endpoints that fall in the ijth cell, and m_{ij} is the number of endpoints for the same cell with pollutant concentrations higher than the set criterion value. The PSCF values should be weighted according to n_{ij}. In this study, the weighting function (W_{ij}) was defined as follows:

$$W_{ij} = \begin{cases} 1.00 & 10 \times \overline{n_{ij}} < n_{ij} \\ 0.70 & 5 \times \overline{n_{ij}} < n_{ij} \leq 10 \times \overline{n_{ij}} \\ 0.40 & 2 \times \overline{n_{ij}} < n_{ij} \leq 5 \times \overline{n_{ij}} \\ 0.05 & n_{ij} \leq 2 \times \overline{n_{ij}} \end{cases} \quad (2)$$

where $\overline{n_{ij}}$ is the mean n_{ij} value. In this study, a potential source analysis was conducted for the nucleation and accumulation mode particles, which represented the air mass influence on the NPF event and the particles from long-range transport, respectively. The criterion values of $PM_{2.5}$ and PM_{10} mass concentration were 75 μg cm^{-3} and 100 μg cm^{-3}, which was the mean value in March, April and May in 2021.

The PSCF results (Fig. S7) showed that high $PM_{2.5}$ mass concentration at CAMS was dominated by two sources, the northwesterly and westerly originating air mass containing dust particles, and the southerly air mass with high mass loading of anthropogenic aerosols. However, for PM_{10} mass concentration, the high values only contributed by the air masses passing through Inner Mongolia and carrying dust particles.

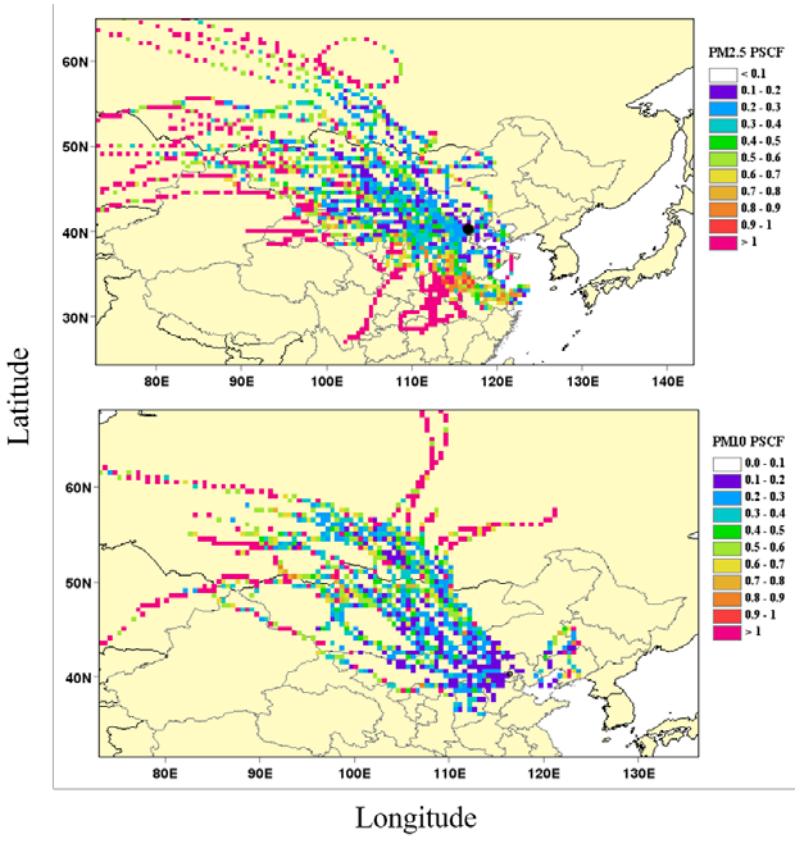

Fig. S7. Air mass classification of back trajectories arriving at the CAMS site in

March, April and May, 2021. The color bar indicates the number concentration weighted potential source contribution function (PSCF) value of (upper panel) $PM_{2.5}$ and (lower panel) PM_{10} mass concentration.

Ashbaugh, L.L., Malm, W.C., Sadeh, W.Z., 1985. A residence time probability analysis of sulfur concentrations at Grand Canyon National Park. Atmospheric Environment 19 (8), 1263–1270.

- How were the dust events defined, not the distinction between the dust episodes, but the identification itself, was it based on airmass trajectories?

Reply: we give the definition in the section of 3.1 with original line 143-145. Three types of dust days are classified based on visibility (National Weather Bureau of China, 1979; Wang et al., 2005), including dust storm with visibility below 1.0 km; blowing dust with visibility of 1.0-10 km and floating dust with visibility below 10 km. The dust days we discussed in the manuscript were identified based on the above algorithm. In this study, the visibility data are from the national surface meteorological observation stations of China Meteorological Administration (CMA). Furthermore, the daily weather phenomena and visibility are issued by CMA (http://www.asdf-bj.net/publish/observation/5.html, last access on 23 March, 2023), which can also help to recognize the dust event. We conducted the identification of dust days during 2017-2021 spring based on the visibility data. We have supplemented the visibility data origins in the manuscript.

The 72 hours back trajectories on dust days March 15-17, 2021 were also calculated and given in the supplementary materials (Fig. S2). It showed the air mass passing through Mongolia, which is the source of dust storm.