

Response to Referee #1

We greatly appreciate the helpful comments from the reviewer, which have helped us improve the paper. We have addressed the comments carefully, as detailed below.

1. An urgent task facing the Chinese government and the scientific community is to quantify the sources and formation mechanisms causing episodic events of high $PM_{2.5}$ mass concentrations and severe haze. This paper provides a summary of source apportionment studies on haze events, but not on $PM_{2.5}$ mass concentrations. It is recommended to also include a review of source factors identified for $PM_{2.5}$ in various regions of China.

Response: We have found more than 40 SCI articles on $PM_{2.5}$ source-apportionment studies published during 2000-2017. We have provided a summary table (Table S2) of these studies in the SI document, and added a new section (2.3) in the revised paper focusing on source-apportionment studies. In this section, we first briefly summarized common receptor models used in $PM_{2.5}$ source-apportionment studies and common source factors found in Chinese cities. We then discussed annual and seasonal contributions of dominant source factors to $PM_{2.5}$ mass region by region.

In the abstract, we have also provided a summary of major findings based on the review of these studies, which reads: "Source apportionment analysis identified secondary inorganic aerosols, coal combustion, and traffic emission as the top three source factors contributing to $PM_{2.5}$ mass in most Chinese cities, and the sum of these three source factors explained 44% to 82% of $PM_{2.5}$ mass across China. Biomass emission in any cities, industrial emission in industrial cities, dust emission in northern cities, and ship emission in coastal cities are other major source factors, each of which contributed 7-27% to $PM_{2.5}$ mass in applicable cities.

2. For a few cities such as Beijing, Shanghai and Guangzhou, inter-annual variations are discussed based on field measurements conducted by different researchers (and likely using different instruments and/or QA/QC methods). How much confidence do you have on these inter-annual variations compared to measurement uncertainties?

Response: We have carefully collected the information about the measurement and analysis methods used in literature and identified potential measurement uncertainties for the dominant chemical components (OC/EC and water soluble inorganic ions). We have added this information in section 2.2 in the revised paper:

"To ensure the comparability of the data collected using different instruments, measurement uncertainties were first briefly discussed here. Most studies in China analyzed OC and EC using DRI carbon analyzer or Sunset carbon analyzer. IMPROVE is the most widely used thermal/optical protocol for OC and EC analysis for DRI analyzer while NIOSH is the one for Sunset analyzer. OC and EC measured

by the two analyzers are comparable if using the same analysis protocol. For example, Wu et al. (2011) showed that OC from Sunset analyzer was only 8% lower than that from DRI analyzer, while EC was only 5% higher. However, when using different protocols by the two analyzers, the differences were much larger, e.g., EC from NIOSH was almost 50% lower than that from IMPROVE (Chow et al., 2010; Yang et al., 2011a). Note that OC and EC were also measured using a CHN elemental analyzer in 2001-2002 in Beijing, which protocol was similar to NIOSH (Duan et al., 2006). In any case, the measurement uncertainties of total carbon (TC, the sum of OC and EC) were less than 10% (Chow et al., 2010; Wu et al., 2011).

The ions including SO_4^{2-} , NO_3^- and NH_4^+ were measured by ion chromatograph. Measurement uncertainties should be less than 15% in most cases under strict QA/QC procedures (Orsini et al., 2003; Trebs et al., 2004; Weber et al., 2003), but could be larger for ammonium nitrate (NH_4NO_3) since it can evaporate from the filters before chemical analysis under high temperature and low relative humidity (RH) conditions, and this applies to both quartz fiber filter and Teflon filter (Keck and Wittmaack, 2005; Weber et al., 2003). The loss of NO_3^- due to evaporation was found to range from 4% to 84% depending on ambient temperature (Chow et al., 2005). Although the exact magnitudes of measurement uncertainties cannot be determined for NO_3^- and NH_4^+ , they are expected not to affect significantly the inter-annual variations discussed below for the three cities (Beijing, Shanghai, and Guangzhou) considering the small year-to-year temperature changes.”

We have taken into account the above information when discussing the trends of measured species through this section.

3. A related question to question 2 above: is it possible to compare the trends identified in this study to other sources such as the online $\text{PM}_{2.5}$ data, the AOD trend analysis data, or available literature?

Response: As noted in a recent paper by Fontes (2017): “The long trends of $\text{PM}_{2.5}$ concentrations were not fully investigated in China, in particular the year-to-year trends and the seasonal and daily cycles.” They analyzed $\text{PM}_{2.5}$ data from 1999-2008 at five megacities in China. We have added this reference in the revised paper. The data set we collected in this review paper covered much longer periods and all the sites across China.

We have added a brief discussion on the relationship between AOD and $\text{PM}_{2.5}$ at the beginning of Section 3, which reads: “Satellite retrievals of AOD have been widely applied to estimate surface $\text{PM}_{2.5}$ concentrations using statistical models (Liu et al., 2005; Hu et al., 2013; Ma et al., 2014; Wang and Christopher, 2003). Although the correlation between AOD and $\text{PM}_{2.5}$ mass concentration depends on many factors, such as aerosol size distribution, refractive index, single-scattering albedo, and meteorological factors (Che et al., 2009; Guo et al., 2009b; Guo et al., 2017), the

predicted $PM_{2.5}$ mass from satellite AOD data compared well with ground-level measurements (Ma et al., 2014; Xie et al., 2015b). Moreover, the spatial distributions of AOD measured using sun photometers mostly agreed with those retrieved from satellite data (Che et al., 2014; Che et al., 2015; Liu et al., 2016b; Pan et al., 2010).”

4. Please also add sub-section titles in the content lists.

Response: We have added sub-section titles in the contents list.