

Response to the comments

Reviewer one

Atmospheric particulates (aerosols) has significant influence on air quality, human health and regional climate changes. Large uncertainties in estimating the aerosol direct radiative forcing exist due to the uncertainties in the aerosol optical properties which were related to the aerosol emissions, profiles, compositions and mixing states. Thus, it's very important to figure out the temporal and spatial distributions of the aerosols in the regions with high aerosol loadings to better accessing their radiative forcing and regional/global climate effects. This study, which examines the spatial and temporal variations of the PM₁₀, PM_{2.5} and PM₁ over 24 sites in China by using a 9 years near continuous PM data measured by GRIMM180 instruments, is important to some extent and the paper has potential. I recommend the manuscript being published in the journal after the revision listed below being addressed.

Comments:

1. Re-typesetting the manuscript. Also, the manuscript still remains poorly written throughout the whole manuscript and requires many corrections.

Response: The manuscript has been edited by a native English-speaking expert at LucidPapers (<http://lucidpapers.com/>). A certification is attached.

2. Page 15320, line 9 and line 11: It should be “the ratio of PM_{2.5} to PM₁₀” and “the ratio of PM₁ to PM_{2.5}”.

Response: It has been revised to “The ratios of PM_{2.5} to PM₁₀ showed a clear increasing trend from northern to southern China, because of the substantial contribution of coarse mineral aerosol in northern China. The ratios of PM₁ to PM_{2.5} were higher than 80% at most stations”.

3. Page 15320, line 16-17: The authors should show the readers what they found while just what they did.

Response: The brief conclusion was added at the end of the abstract: “Bimodal and unimodal diurnal variation patterns were identified at urban stations. The investigation of meteorological factors effects reveals the emission variation possibly dominates the long-term PM concentration trend; meanwhile meteorological factors play a leading role during a short period”.

4. Page 15323, line 9: The authors indicate the uncertainty of GRIMM in (Grimm and Eatough) was 9.9%, how about in China? Is it also 9.9%?

Response: The comparison result of hourly average PM_{2.5} concentrations simultaneously monitored by GRIMM and TEOM at SDZ (around Beijing, China) in February 2010 shows the good linear relationship between GRIMM and TEOM PM_{2.5} concentrations (Zhao et al., 2011) (Fig. 1). Also an early study in Beijing downtown during the summer 2004 concluded that GRIMM measurements have shown to reproduce very well all the TEOM-FDMS variations (wet and dry periods) suggesting that optical measurements could be used derive PM_{2.5} and could also account for semi-volatile material in aerosols (Sciare et al., 2007). There is no specific uncertainty value given by above studies, and such uncertainty value is not constant in different regions and periods. The point is to give the reference evidences that the GRIMM data are acceptable for PM measurement compared with traditional used instruments such as TEOM.

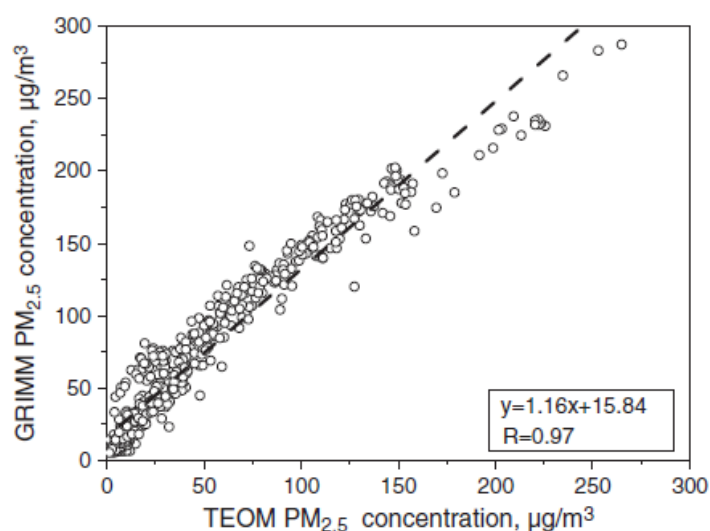


Fig. 1. GRIMM vs. TEOM PM_{2.5} concentration, 1h average, SDZ, February 2010 (Zhao et al., 2011)

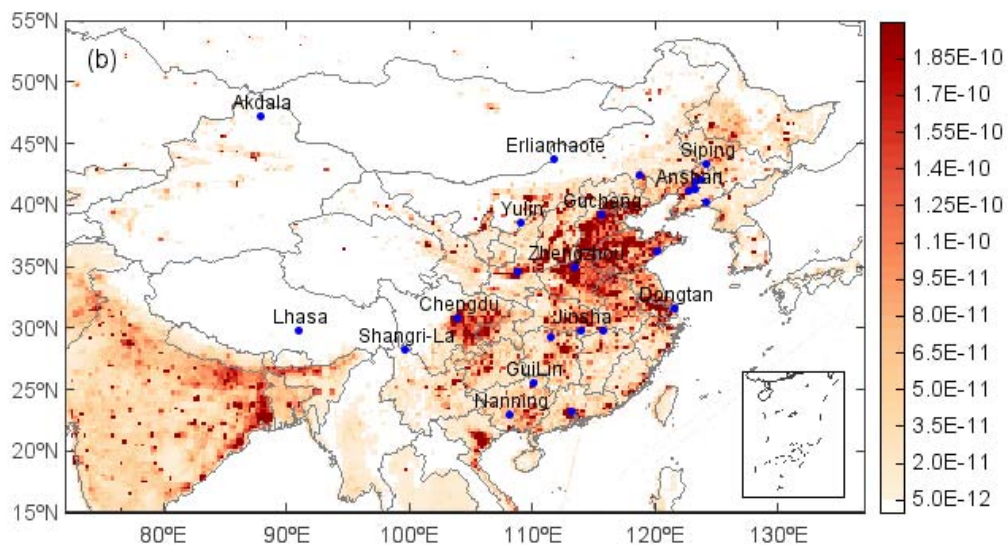
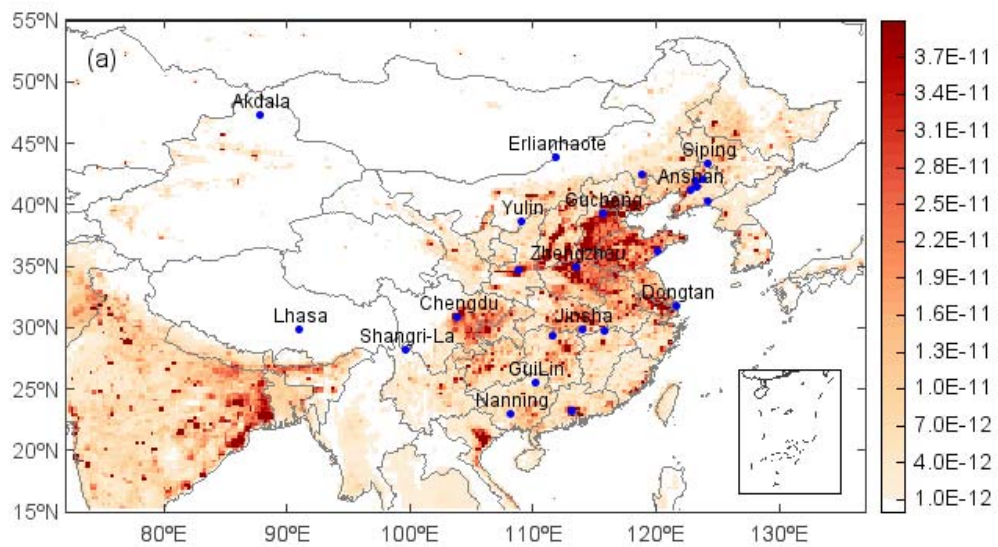
The above two references has been added in revised manuscript: “The GRIMM measurement in Beijing, China have shown the good linear relationship with TEOM and suggesting that optical measurements could be used derive PM_{2.5} and could account for semi-volatile material in aerosols (Sciare et al., 2007; Zhao et al., 2011)”.

5. Page 15324, line 1: The authors should present some emission results in China form publications (e. g. Q. Zhang et al., 2009) when explaining the reasons.

Response: Some emission results in China have been added in revised manuscript. Detailed description could be found in below response.

6. Again, it's obvious that the results being analyzed is too simple. The authors should provide evidences (from the similar studies in publications/references or from self-analysis) to make the readers more clearly.

Response: We have enhanced the analysis part with especially more emission results in China. In revised manuscript, section 3.6 was changed to “Emission and meteorological influences” with emission data analysis based on HTAP harmonized emissions database (<http://iek8wikis.iek.fz-juelich.de/HTAPWiki/WP1.1>). Anthropogenic emission distributions of BC, PM_{2.5}, SO₂ and NO₂ in 2010 were presented in figure 2 with similar spatial pattern. PM loadings in China were generally similar to this emission pattern. For example, most PM pollutant stations located in highest emission region of HBP. Therefore, PM loadings were controlled by anthropogenic emission amount in mid-east China.



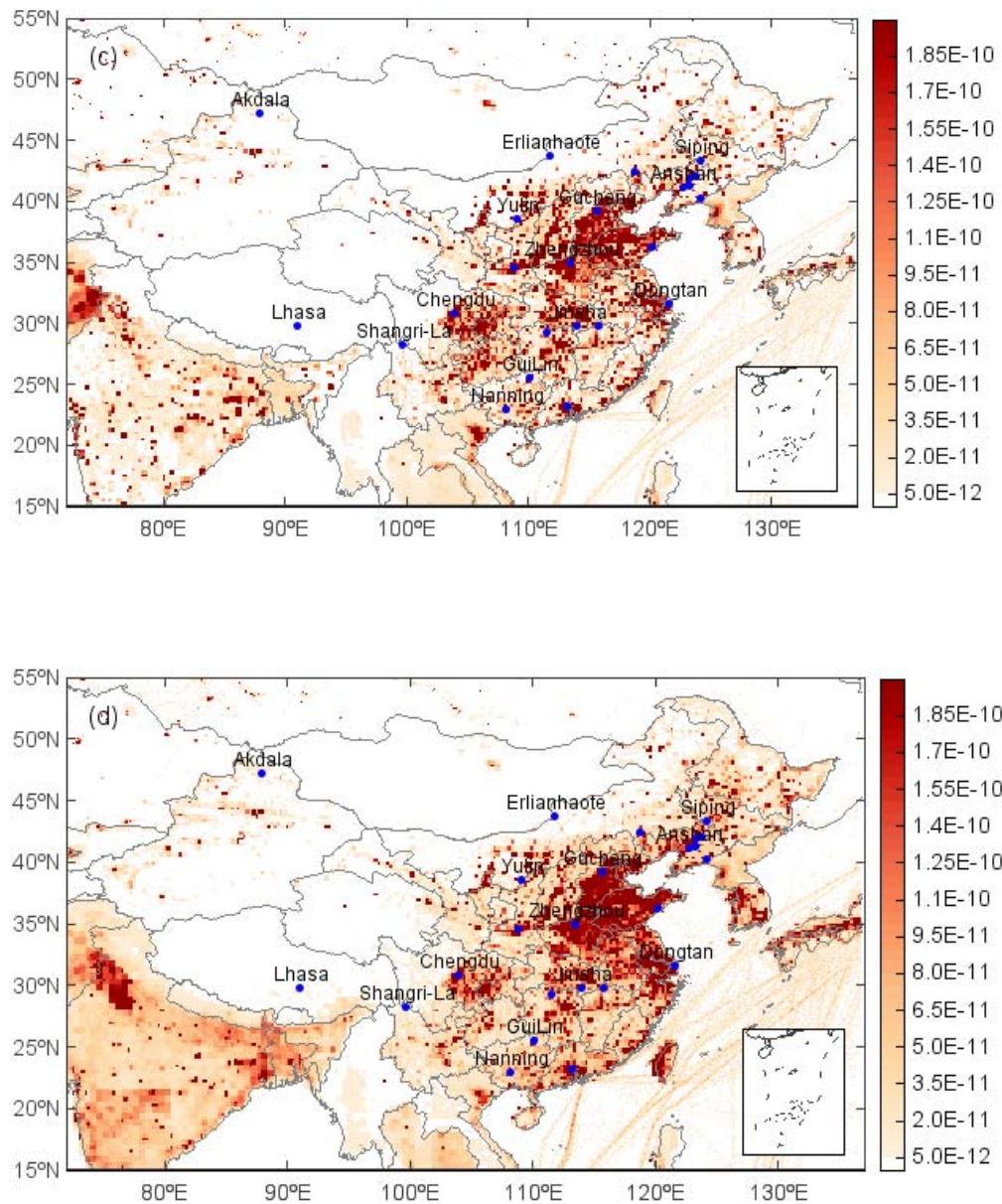
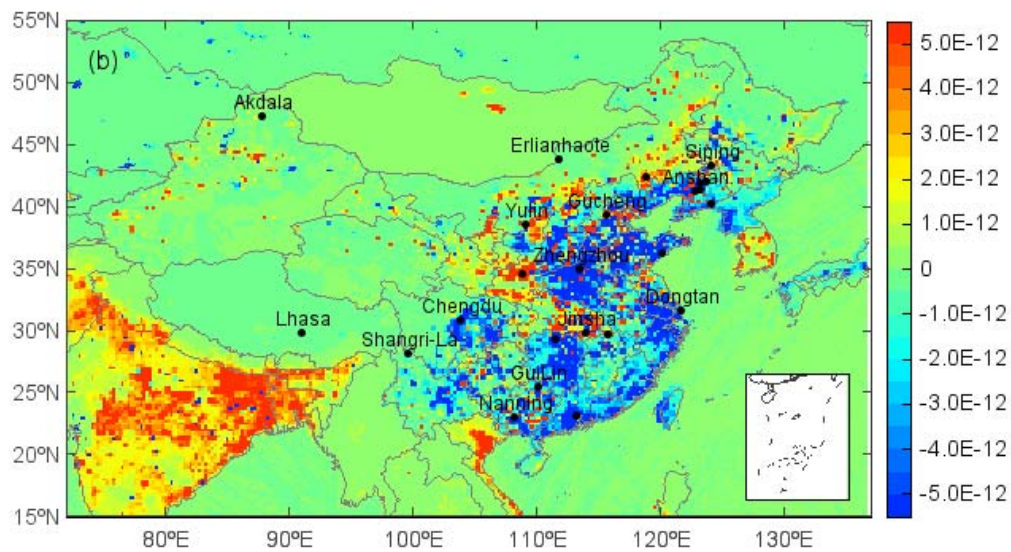
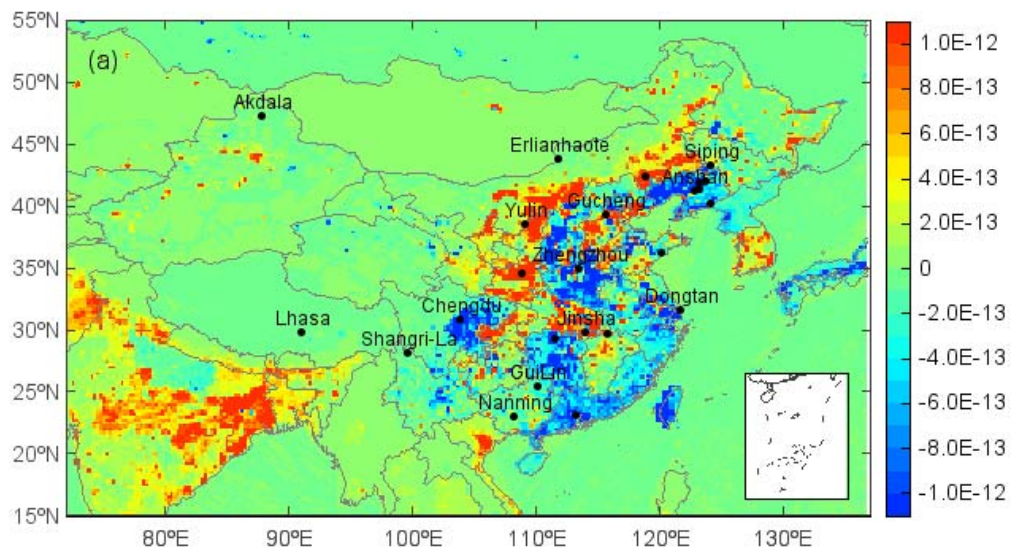


Fig. 2. Anthropogenic emission distributions at 0.1 degree \times 0.1 degree resolution of (a) BC, (b) PM_{2.5}, (c) SO₂ and (d) NO_x (units: kg m⁻² s⁻²) based on HTAP_v2 dataset

The emission trends in China during 2005-2010 (Wang et al., 2014) were also presented to explain the long-term PM trends. The emissions of SO₂ and PM_{2.5} in East Asia decreased by 15 and 12 %, meanwhile the emissions of NO_x and NMVOC increased by 25 and 15 % (Wang et al., 2014). Also spatial distributions of emission difference between 2010 and 2008 were studied using HTAP_v2 emission dataset (Fig. 3), so the PM vs emission variation in different regions could be studied. Although there is no published emission data after 2010, it was believed that the emission was greatly controlled after end of 2013 with the issue of “Action Plan for the Control of Air Pollution” document. It can explain the general decrease trend in

2014. The conclusion of emission influence on long-term PM pattern could be supported from above analysis. More detailed description was added in revised manuscript.



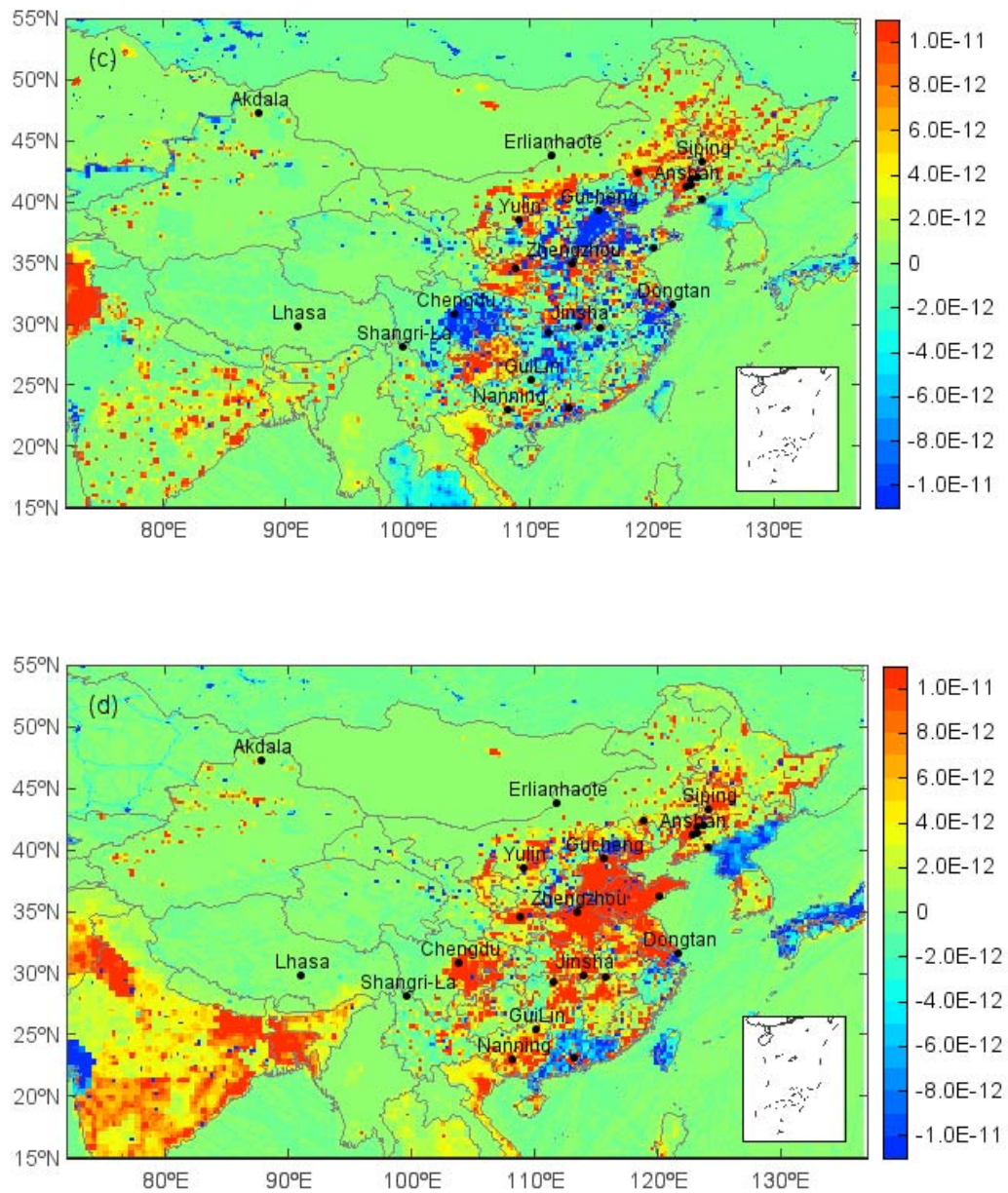


Fig. 3. Emission difference between 2010 and 2008 at 0.1 degree \times 0.1 degree resolution of (a) BC, (b) PM_{2.5}, (c) SO₂ and (d) NO_x (units: kg m⁻² s⁻²) based on HTAP_v2 dataset

7. In addition to diurnal and seasonal cycles, did the PM have any periodic in China.

Response: There is no obvious general long-term periodic of PM in China from this 9-year data study.

Reviewer two

This manuscript presented PM concentrations over China measured by China Atmosphere Watch Network. Although PM_{2.5} measurements are available over China since 2013, the measurement data presented in this paper are valuable because it covers longer period (2006-2014) and provides PM₁/PM_{2.5}/PM₁₀ information. This dataset would be very helpful for understanding the evolution of PM concentration over China. The manuscript could be published in ACP after the following concerns are addressed.

Comments:

8. Sect. 2. The authors claimed that GRIMM measurements are in good agreement with TEOM. I am curious if the authors compared GRIMM with other instruments under heavy pollution condition in China. Referring to comparison in western countries is not very convincing.

Response: This comment has been answered in comment 4.

9. P15326, L6-16, more comparison for the same period (2006-2014) would be meaningful.

Response: More comparison for the same period (2006-2014) has been added in the revised manuscript.

“The average PM₁₀ and PM_{2.5} concentrations were 23.9 and 16.3 $\mu\text{g m}^{-3}$ for the period 2008 – 2009 in Netherlands (Janssen et al., 2013). The 20 European study areas observation results from ESCAPE project between October 2008 and April 2011 showed PM₁₀ and PM_{2.5} have similar spatial pattern with low concentrations in Northern Europe and high concentrations in Southern and Eastern Europe (Eeftens et al., 2012)”

10. P15328, Sect. 3.4, this could be the most important section in the manuscript. In this section, inter-annual variations of PM_{2.5} concentrations of individual sites are presented one by one. It would be very boring the international readers who don't familiar with Chinese cities. The overall PM trend over China and the driven forces behind the trend are missing in this section.

Response: There is no overall PM trend over whole China except the decrease trend in 2014, so the trend analysis was done in different areas of China, such as HBP, northeast China, southern China and so on. Such summary sentence has been added in revised manuscript. The driven forces behind the trend were analyzed by emission and meteorological analysis in section 3.6. (See response of comment 6)

11. P15330, Sect. 3.5, I would like to see diurnal variations in PM₁ and PM₁₀ concentrations and if they are similar to PM_{2.5}.

Response: The diurnal variations in PM1 and PM10 concentrations are similar to PM2.5 (Fig. 4). An example figure for stations of Zhengzhou, Xian and Gucheng is given below. So we add a sentence of “The diurnal variations in PM₁ and PM₁₀ concentrations are similar to PM_{2.5} at most stations” in revised manuscript.

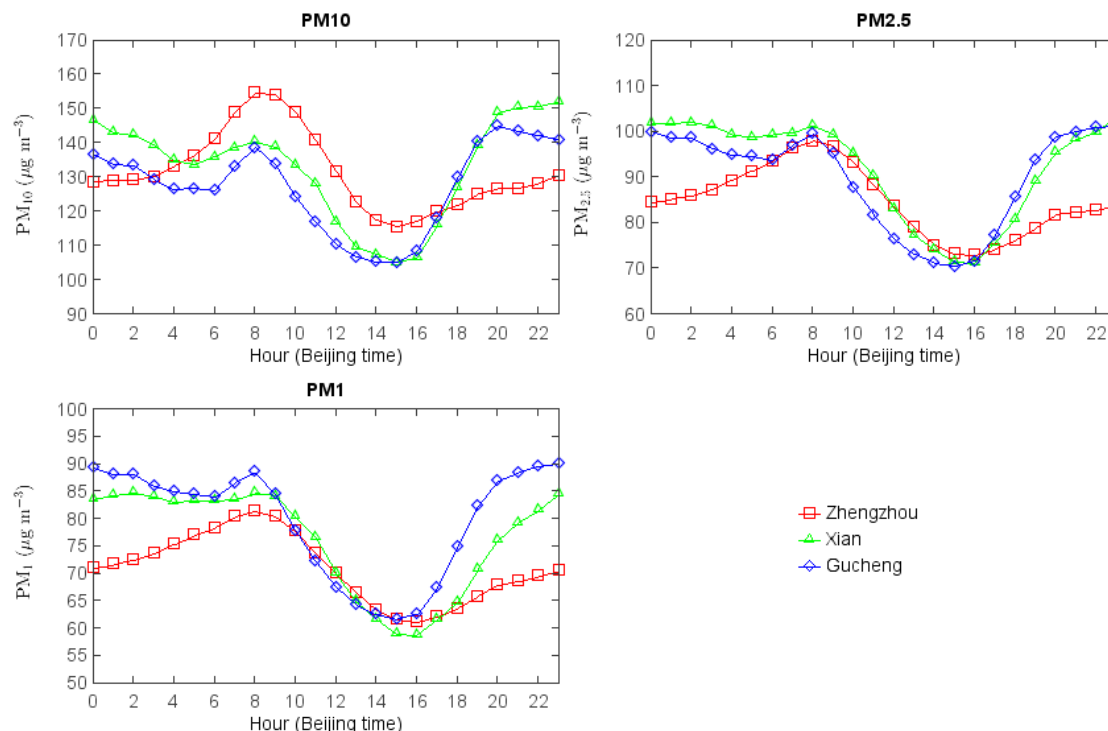


Fig. 4. Diurnal variations in PM10, PM2.5 and PM1 at Zhengzhou, Xian and Gucheng

12. P15332, L16-17, the authors concluded that “emission variation must to be considered for long-term trend analysis especially in rapid developing countries.” Emission data should be used in the discussion to support their arguments.

Response: The emission variation data have been provided for this analysis. Please see the response to comment 6.

13. Figures. Fig. 1 and Fig. 2 could be combined into one figure.

Response: Fig. 1 and Fig. 2 were combined as below one figure.

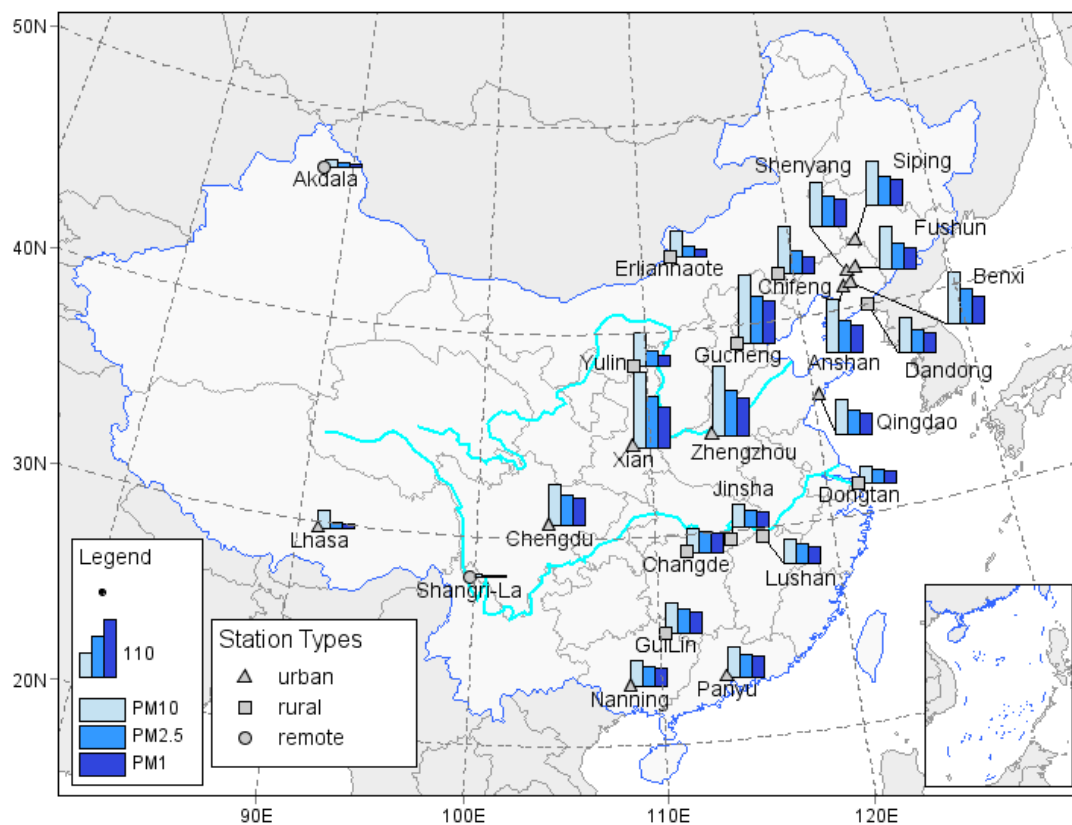


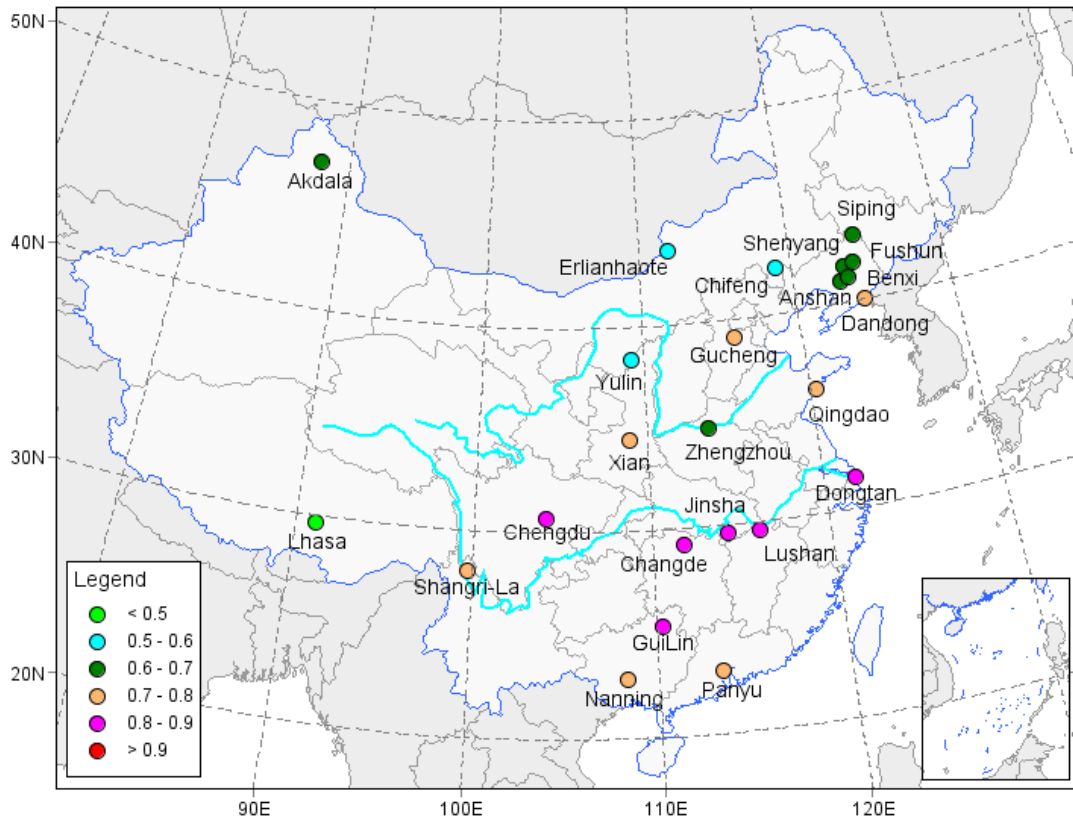
Fig. 5. Map showing PM observation stations and their bar charts of average PM₁₀, PM_{2.5} and PM₁ concentrations ($\mu\text{g m}^{-3}$)

14. Figure 3 could be removed because it doesn't provide additional information than Table 2.

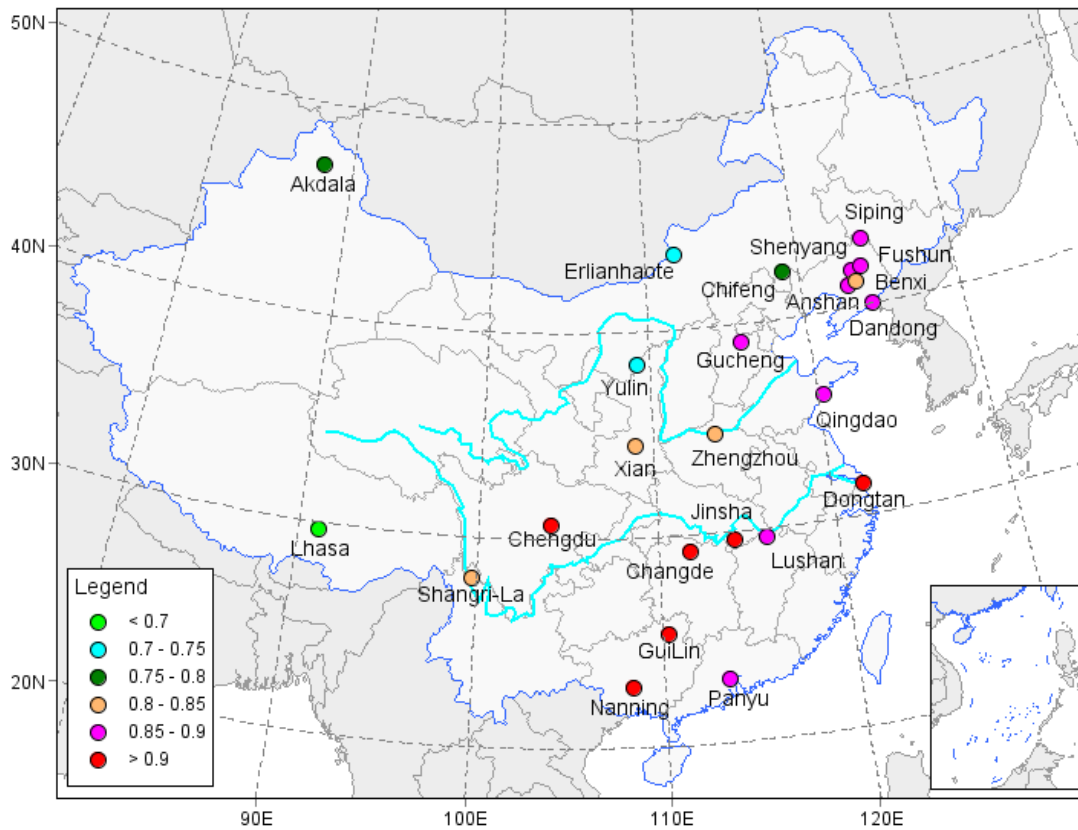
Response: We agree and have removed figure 3 in revised manuscript.

15. PM₁/PM_{2.5} ratios should be also presented in Figure 5.

Response: PM₁/PM_{2.5} ratios have been presented in Figure 5 as below.



(a)



(b)

Fig. 6. Spatial distribution of the average ratios of (a) $PM_{2.5}/PM_{10}$ and (b) $PM_1/PM_{2.5}$.

Reference

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- Zhao, X., Zhang, X., Pu, W., Meng, W., and Xu, X.: Scattering properties of the atmospheric aerosol in Beijing, China. *Atmos. Res.* **101**, 799-808, 2011.