

Response to Referee #1:

Interactive comment on “Atmospheric wet and dry deposition of trace elements at ten sites in Northern China” by Y. P. Pan and Y. S. Wang

Anonymous Referee #1

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General Comments:

The authors report measurements of atmospheric dry and wet deposition of trace elements in ten sites in the North China Plain area which have experienced serious air quality problems associated with intensive agricultural and industrial activities. The results of this paper are quite interesting.

The authors investigated the relative importance of wet and dry deposition in the removal of airborne trace elements and combined contribution of wet and dry deposition to ecosystems in the region. This will help researchers improve the understanding of the transport of air pollutants from sources to the environment. The findings are important for model validation and are also helpful for policy makers to create legislation to reduce the emissions and protect soil and water from air pollution. Overall, the paper is well presented and straight-forward. I can recommend publication, once the authors address the comments below.

Response: The authors appreciate the valuable suggestions given by Referee #1 for improving the overall quality of the manuscript. In the revised version we have made the corresponding corrections according to the reviewer's comments. Below are the responses to every comment one by one.

Page 20648, Line 20-25: Should the author give the full name when he/she first mentions the chemical species? Please carefully check the whole manuscript.

Response: We agree with the suggestion above. We have checked through the text to give the full name when a chemical species appeared for the first mention.

Page 20651, Line 19: In the results and discussion part, the author talked about TEs related to local emissions. Could the author add a paragraph in the introduction to briefly introduce the sources of TEs in the atmosphere? What TEs were from human activities and what TEs were from natural sources?

Response: [We have incorporated the suggestions and added the source information of airborne TEs in the third paragraph in the Introduction.](#)

Page 20651, line 24: It would be better to move figure S1 in the Supplement to the text for the readers' convenience.

Response: [The suggestion is implemented.](#)

Page 20652, Line 15-20: I noticed this automatic collector was only used in the authors' research group. If this sampler has been used in other studies or in different research groups, please cite one or two of their papers.

Response: [The automatic collector was produced by a custom company \(APS-2B, Xianglan Scientific Instruments Co., Ltd., Changsha, China\) and has been used in South China by other research groups \(\[Huang et al., 2010\]\(#\); \[Wang et al., 2010\]\(#\); \[Wang et al., 2014\]\(#\); \[Zhang et al., 2011\]\(#\)\), but most of them were used to collect wet deposition only. We have added these papers in the citation.](#)

[Based on the automatic sampler we developed a unique technique sampling dry deposition using a PUF filter combined with a glass bucket. The method was optimized and verified by the national standard method in this study. More details were added in the revised paper.](#)

Page 20653, Line 10-15: If the authors used the same sample treatment procedures as others, please cite one or two papers.

Did the author conduct duplicate sampling such as setting up two samplers at one site during the measurements? If the author did this, please include the data in the manuscript. If not, I highly recommend thinking about this in future studies.

Response: The sample treatment procedures were established in our previous experiments and reported elsewhere ([Pan et al., 2012](#), [2013b](#); [Pan et al., 2010](#)). We will cite the reports as suggested.

To check the quality of the sampling, different samplers were used to collect precipitation during the site-maintenance visits at the initial stage of the experiment at the BJ and CZ sites. The results showed no significant difference between the samplers for most of TEs. In the revised paper the comparison of two samplers for selected TEs were shown in a figure.

Also at the BJ and CZ sites, the PUF-based bucket used as a dry deposition collector was further evaluated and compared to the standard method recommended by the Ministry of Environmental Protection of China, which uses buckets containing glycerol as an alternative technique. The results observed for the two methods agreed well with one another and more discussions were added in the revised manuscript.

Page 20656, line 21: I suggest the authors move Table S1 (also S2 and S3) in the Supplement to the text for the readers' convenience. Otherwise, please add the 10-site mean values measured in this study in these tables for comparison.

Response: Thanks for this comment. We have moved Table S1, S2 and S3 in the Supplement to the revised manuscript.

Page 20659, Line 18: Dust storms are a regional transport episode, but the author only mentioned impacts at the BJ site. Did the author find any evidence indicating impacts of dust storms on the other sites in this region?

Response: We are sorry for this confusion. With the exception of BD and TS, most sites in this study suffered from the regional transport of natural dust, especially during spring. To confirm that, we have checked the Sand-Dust Weather Almanac issued by the Chinese Meteorological Administration and found that there were 31 sand-dust weather events recorded in China between 2008 and 2010. Of the total, 16 events reached the target regions during the period (9 events occurred in spring); all of which were blowing or floating dust and no sand-storms were recorded. We thus

conclude that the long-range transport of natural dust from the northern/northwestern deserts and loess deposits results in the relatively high dry deposited elemental flux in spring than in other seasons in this study. In addition, sand-dust weather events decreased eastward due to the effects of distance and particle size. As a result, there were more days with blowing or floating dust at BJ than east coastal site of TG, according to the recorded weather phenomena. Dry deposition of Al at these two sites (2.1 and 1.3 g m⁻²) during spring also supported this phenomenon.

The above discussions were briefly summarized in the revised paper.

Page 20659, Line 23: Beijing is also a coal consumption city during the winter. Did the author find evidence of this at the BJ site? If not, please explain why.

Response: We agree with the reviewer that coal is still the primary fuel in Northern China and widely used for industrial processes and daily life, and more coal is combusted for heating in winter. Consequently, dry depositions in the cold seasons were expected to be enhanced in the region where a great deal of coal was combusted. This is supported by the elevated flux of various TEs at the urban and industrial sites of TJ, BD and TS, compared with other sites. In the urban areas of Beijing, however, the energy used for heating and industrial processes was mainly electricity and natural gas except for limited residential coal consumption ([Zhao et al., 2013](#)). At present, annual coal consumption in Beijing was about 21 million tons, which is significantly lower than that in Tianjin and Hebei (70 and 300 million tons). As a consequence, the dry deposition of coal combustion related TEs (e.g., Pb and Tl) in BJ was lower than that in TJ, BD and TS, but still higher than that in YC, CZ and XL, indicating the influences of residential coal consumption in the urban areas of Beijing.

In the past ten years, with the gradual replacement of coal by natural gas and electricity in urban Beijing, the sulfate and elemental carbon in winter decreased gradually from 25 and 8.7 µg m⁻³ to 14 and 6.3 µg m⁻³, respectively ([Zhao et al., 2013](#)). Further decrease of elemental deposition in Beijing can be expected, if the reduction of coal consumption continues.

We have revised the manuscript to reflect the above discussions.

Page 20660, Line 19: If the wdfPb was much higher than other places, please explain what are the major sources of wdfPb in this region?

Response: Besides natural sources from regional and local soil, possible anthropogenic sources of Pb include coal combustion, vehicle exhaust, cement factories and smelters ([Cheng and Hu, 2010](#)). But the relative contribution of the above sources is of spatial and temporal variable. After the phase-out of leaded gasoline in China since 2000, the major emission sources of airborne Pb in eastern and central China were estimated to be coal consumption and non-ferrous metal smelting, instead of vehicle exhaust ([Li et al., 2012](#)). However, detailed Pb isotopic signatures of PM₁₀ from selected sites in North China indicated its source was mainly anthropogenic, and mostly attributable to coal combustion and vehicle emissions with additional industrial sources ([Luo et al., 2014](#)). A case study in Beijing found that airborne Pb predominantly from anthropogenic sources was reduced by approximately 50% during the 2008 Olympic Games due to the mitigation measures implemented by the Chinese Government ([Schleicher et al., 2012](#)). Moreover, the temporal variations of Pb concentration correlated to the restrictiveness of relative measures, especially during different traffic restrictions, further demonstrating the significance of traffic sources ([Chen et al., 2014](#)). But the vehicular emissions from urban areas (e.g., Beijing) are not likely an important regional source of Pb and thus have insignificant impacts in rural areas (e.g., Xianghe) ([Li et al., 2010](#)). We conclude that Pb in wet deposition on the regional scale is mainly emitted from industrial processes and coal burning. These emissions can be widely dispersed throughout the atmosphere and transported to the downwind regions ([Zhao et al., 2013](#)), resulting in the high wet depositions at the background site of XL (discussed in Sect. 3.2.3).

Page 20662, Line 7: Is there any report of long-range transport effects on wet deposition flux of TEs? The author should add more discussion on this point. The author only mentioned “upwind areas”. Is it long-range transport from urban and industrial areas or rural areas?

Response: Yes, the long-range transport effects on wet deposition flux of TEs were well recorded. For example, wet deposition fluxes of TEs measured along the Japan Sea coast have been strongly affected by the long-range transport of air pollutants from the Asian continent during winter and spring ([Sakata et al., 2006](#)). A recent study also found that long-range transport of pollutants from south Asia has a significant impact on the trace elements in atmospheric wet deposition in the high altitude remote areas in the southern slope of the Himalayas ([Tripathy et al., 2014](#)).

Since the emissions of industrial pollutants and fossil fuel combustion from upwind sources in Tianjin and Hebei are prominent, trace elements in precipitation observed at XL could be from regional emission sources. Imprints of regional transport were indicated by the fact that the metallic episodes experienced at the XL site closely associated with the air mass from southeast that passed TS and TJ, or from southwest that passed BD and SJZ ([Pan et al., 2013a](#)).

As suggested we have added more discussion on the issue of long-range transport.

Page 20664, Line 15: Please cite the paper which partially verified this pattern in Northern China.

Response: We are sorry for this confusion. In the revised version, “Northern China” was changed to “in this study”. That is, this pattern was partially verified in this study.

Page 20667, line 3: I suggest the authors add a figure to show the vertical profile showing the distribution of M_c for each metal vs. soil depth, which will be helpful for the readers to interpret the ideas examined here.

Response: We accept this suggestion and have added a figure in the revised paper.

Page 20667, line 27: Also, it is better to move Table S4 in the Supplement to the text for the readers' convenience.

Response: Done.

Page 20674, Table 1: It would look better if the author can add several black lines in

Table 1 to clearly separate the urban, industrial, suburban, agricultural and rural groups.

Response: [The suggestion is implemented.](#)

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