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Interactive comment on "Distribution and Sources of Air pollutants in the North China Plain Based on On-Road Mobile Measurements" *by* Yi Zhu et al.

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We thank the reviewer for the critical comments, which are very helpful in improving the quality of the manuscript. We are revising the manuscript according to the suggestion and the following lists our responses to the comments.

Referee Comment #1:

This manuscript reports measurements of SO2, NOx, CO and black carbon made in a mobile van traveling on five expressways in the North China Plain in summer 2013. The authors offered some general discussions on sources and long-range transport of these pollutants. On-road measurements are normally used to understand emission characterises of road traffic, but this appears not the case for this study which attempts to study the spatial distributions of the air pollutants in the NCP region. I doubt this

objective can be achieved due to potentially large impact from vehicle emissions on the data. Another concern is that the reported data may have major flaws. The extremely high NOx values are not consistent with moderate values of the other three pollutants. If the NOx data are correct, they (mean value=452 ppbv) clearly show huge impact of on-road vehicles on the measurements. However, the mean CO value is only $\sim$1 ppm, which seems too low. Is this due to the dominance of diesel vehicles on the highways? If so, the measured black carbon would be significantly affected too by the diesel vehicles. How were the instruments calibrated? Did you make measurements off road to compare with the on-road data to check the impact of road vehicle emissions? In summary, the authors are advised to clarify these two important issues (the intended use of the on-road measurements and the data quality). In addition, the analysis and discussion of the data set should be more in-depth.

Response: 1. In previous studies, on-road mobile measurements were not only used for estimating the vehicular emissions, but also for analyzing the spatial distributions and the relationship between cities and regions (Kolb et al., 2004; Johansson et al., 2008; Wang et al., 2011).

The influences of vehicular emissions on on-road mobile measurements were different for NOx, CO and SO2. According to the analyzation in section 3.2 in the paper, NOx was mainly from vehicular emissions. According to the reports of SO2 emission inventory (Li et al., 2015) and vehicular emission inventory (Cai and Xie, 2007), the contribution of vehicular emission to SO2 was not significant during the time of our measurements.

Based on the monitoring results along the highways in our study area (Zhang et al., 2003), the ratio of the amount of gasoline and diesel vehicles was about 1:1. During our measurements, we found that the diesel vehicles were mainly diesel buses and median-duty diesel trucks. With the reported on-road diesel vehicular emission factors for CO and NOx in China (Table 1) (Shen et al., 2015), we calculated that the overall vehicular emission factors for CO and NOx during our measurements were 3 g/km and

5 g/km. The concentration of NOx on road was about 400 ppb higher than that off road. Thus, it is estimated that the concentration of CO on road was about 240 (=400/5*3) ppb higher than that off road, which contributed about 24% of the total concentration. It can also be found from the temporal distributions of concentrations of CO, NOx and BC, e.g. in June 13 (Fig. 1), that the concentration of CO on road (482 ppb) was about 25% higher than that of off-road (391 ppb).

In conclusion, vehicular emissions contributed less than 30% of CO measured in this experiment, and the CO and SO2 measurement results could be used for studying the spatial distributions of the air pollutants in the NCP region.

2. In previously reported studies of on-road mobile measurements, the daily concentration of NOx could be higher than 400 ppb. Considering that the concentration of CO on road was about 25% higher than that of off-road, and the background concentration of CO in the North China Plain was 0.4-0.7 ppm, the result of CO measured in this experiment was reasonable. Besides, the concentrations of NOx and BC measured had similar trends (Fig. 1) and correlated each other well (Fig. 2). Based on this regression curve, the ratio of emission factors of BC and NOx was 0.004:1, which was close to the previously reported diesel vehicular emission factors in Beijing (0.008:1) (Wang et al., 2012). It could be conclude that the diesel vehicular emissions had a large impact on the BC and NOx concentrations measured in NCP.

We have carried out quality assurance and quality control of the on-road measurements, including instruments calibrations and inter-comparison with monitoring station in the campus of Peking University (Wang et al., 2009). One of the instruments calibrations results is shown in Figure 3.

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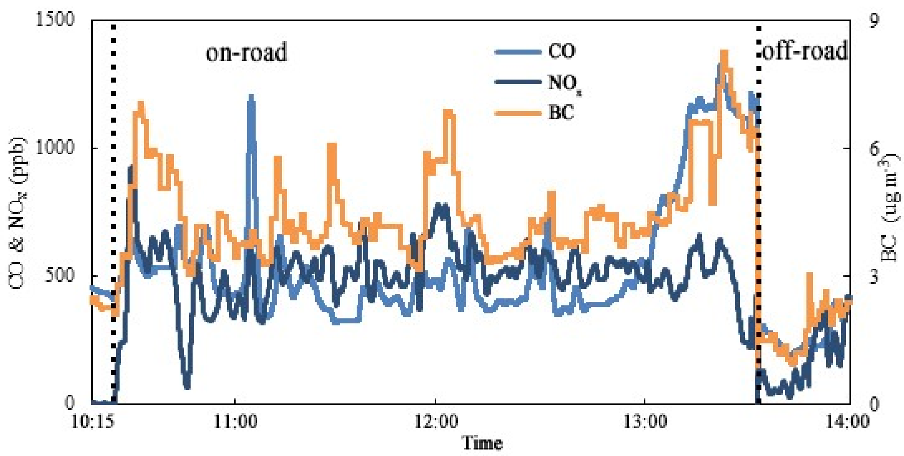

Fig. 1. Temporal distributions of concentrations of CO, NOx and BC in June 13.

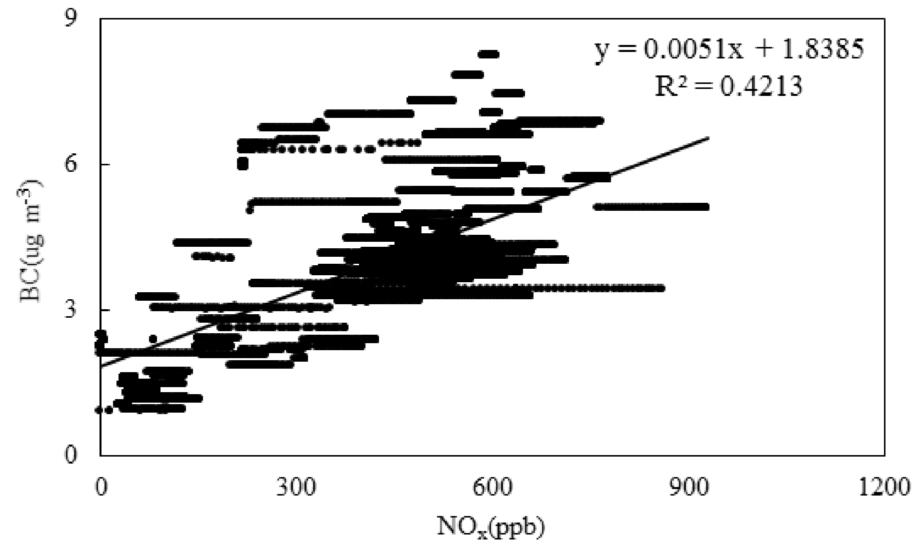

Fig. 2. The regression curve of the concentrations of NOx and BC in June 13.

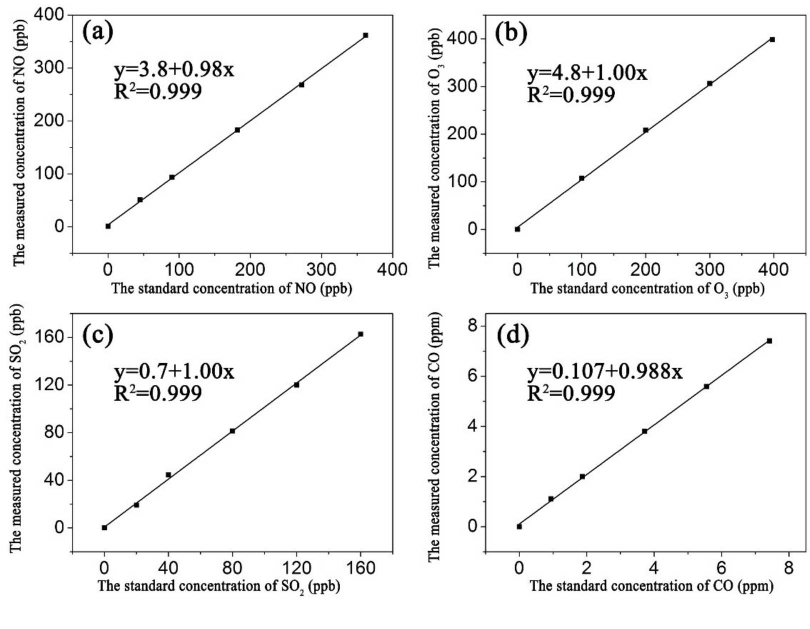

Fig. 3. Calibration curves of gas analyzers in June 16.

Vehicle type	Diesel bus	Median-duty diesel truck	Light-duty gasoline vehicles
CO	1.2 g/km	1.5g/km	4.4 g/km
NO_x	11 g/km	6.4 g/km	1.3 g/km

Fig. 4. On-road diesel vehicular emission factors for CO and NOx in China (Shen et al., 2015).