

Responses to the Comments of the Referees

Distribution and Sources of Air pollutants in the North China Plain Based on On-Road Mobile Measurements

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We thank the reviewers for the critical comments, which are very helpful in improving the quality of the manuscript. We have made major revision based on the critical comments and suggestions of the reviewers. Our point-by point responses to the comments are listed in the following.

Referee #1:

Comment No. 1. This manuscript reports measurements of SO₂, NO_x, 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.

Response: Actually in previous studies, on-road mobile measurements have also been used for analyzing the spatial distributions (Kolb et al., 2004), the urban emissions (Johansson et al., 2008), and the regional transport of air pollutions (Wang et al., 2011).

We agree that we need to provide more evidences to justify the representativeness of our dataset for the regional background concentrations of air pollutants. Following the comments and suggestions of the reviewers, we have rewritten the manuscript and provided the following evidences:

- 1. Comparing the on- and off-road measurements of air pollutants to estimate the enhancement of air pollution on highway above the regional background**

Each on-road measurement trip started from a parking lot in a highway service center and ended at the parking lot of another service center. The parking lots are about 150 m away from highway, such as those in service centers Dezhou (DZ) and Xizhaotong (XZT) (Fig. 1). Using the difference of the concentrations of air pollutants measured in a parking lot and on highway, we can estimate the level of the enhancement of the concentrations of air pollutants on highway above regional backgrounds.



Fig. 1. The locations of the mobile platform parked at the parking lots in the service centers Xizhaotong (XZT) and Dezhou (DZ), and their distances to high ways (from Google Map).

Table 1 shows the 5 min averaged concentrations of NO_x , CO, SO_2 , and BC measured in parking lots and on highways, and the difference between the on- and off-road concentrations. The concentrations in parking lots were measured for 5 mins before driving or after parking with engine turned off, and the concentrations on highways were measured for 5 mins after entering highways or 5 mins before entering the service centers.

The concentrations of NO_x measured on highway show drastic enhancement than those measured off-road, from 19 to 449 ppb, or 43% to 1658% ($510 \pm 61\%$, mean \pm Standard deviation), while other pollutants have much lower enhancement or even reduction in concentrations. The difference between the on-road and off-road concentrations of CO ranged from -478 to 145 ppb, or -28% to 34% ($7 \pm 22\%$); for SO_2 , it is -26 to 13 ppb or -18% to 175% ($52 \pm 59\%$); for BC, it is -0.7 to $4.7 \mu\text{g m}^{-3}$ or -11% to 261% ($85 \pm 90\%$).

The 175% enhancement of SO₂ on July 12 could be due to the much lower SO₂ concentration, 1.2 to 3.3 ppb, than those measured in other days. The rain on July 12 is likely the major reason for the much lower concentrations of SO₂ (3 ppb) and NO_x (63 ppb), while CO and BC, which are much less water soluble, show no significant difference than those measured in other days. If we ignore the results of SO₂ and NO_x on July 12, then the difference between the on- and off-road concentrations were 82% to 1658% (510±61%) for NO_x and were -18% to 87% (31±31%) for SO₂.

Apparently, vehicular emission is the major source lead to the 82% to 1658% enhancement of NO_x concentrations on highway. The enhancement of CO and SO₂ concentrations on highway were mostly less than 30%, suggesting vehicular emission is not the main source for CO and SO₂ on highway, regional background is the dominant factor determine their concentrations.

Table 1. The concentrations of NO_x, CO, SO₂ and BC measured in parking lots and high ways, and the concentration differences between off-road and on-road measurement.

Date	Parking lot	Concentrations							
		NO _x (ppb)				CO (ppb)			
		Off-road (C1)	On-road (C2)	C2-C1	$\frac{C2-C1}{C1}$	Off-road (C1)	On-road (C2)	C2-C1	$\frac{C2-C1}{C1}$
6/11	XZT	234	643	409	175%	694	677	-17	-2%
6/12	XZT	411	699	288	70%	1050	1098	48	5%
6/12	DZ	20	106	86	430%	396	506	110	28%
6/13	DZ	19	334	315	1658%	431	576	145	34%
6/17	XZT	30	297	267	890%	1000	1086	86	9%
6/18	XZT	109	468	359	329%	1212	955	-257	-21%
6/18	DZ	372	677	305	82%	588	711	123	21%
7/3	XZT	49	498	449	916%	826	861	35	4%
7/12	XZT	44	63	19	43%	1708	1230	-478	-28%

Table 1 (continuous). The concentrations of NO_x, CO, SO₂ and BC measured in parking lots and high ways, and the concentration differences between off-road and on-road measurement.

Date	Parking lot	Concentrations	
		SO ₂ (ppb)	BC (μg m ⁻³)

		Off-road (C1)	On-road (C2)	C2-C1	$\frac{C2-C1}{C1}$	Off-road (C1)	On-road (C2)	C2-C1	$\frac{C2-C1}{C1}$
6/11	XZT	11.3	12.7	1.4	12%	2.3	5.6	3.3	143%
6/12	XZT	24.4	30.9	6.5	27%	2.9	6.6	3.7	128%
6/12	DZ	8.9	10.0	1.1	12%	1.5	4.1	2.6	173%
6/13	DZ	14.6	12	-2.6	-18%	2.3	3.6	1.3	57%
6/17	XZT	11	17	6	55%	3.4	5.5	2.1	62%
6/18	XZT	15	28	13	87%	6.6	5.9	-0.7	-11%
6/18	DZ	17	21	4	24%	7.4	10.3	2.9	39%
7/3	XZT	17	21.9	4.9	29%	1.8	6.5	4.7	261%
7/12	XZT	1.2	3.3	2.1	175%	5.3	6.2	0.9	17%

2. Time series of the concentrations of air pollutants measured on highway

Figure 2 shows the concentrations of NO_x, CO, SO₂, and BC measured on highway on a typical day, June 13, 2013. Apparently, the concentration of BC follows the trend of NO_x concentration and those of CO and SO₂ did not. This suggests the high enhancement of NO_x and BC concentration on highway was due to the vehicular emission, while the concentrations of CO, SO₂ were not.

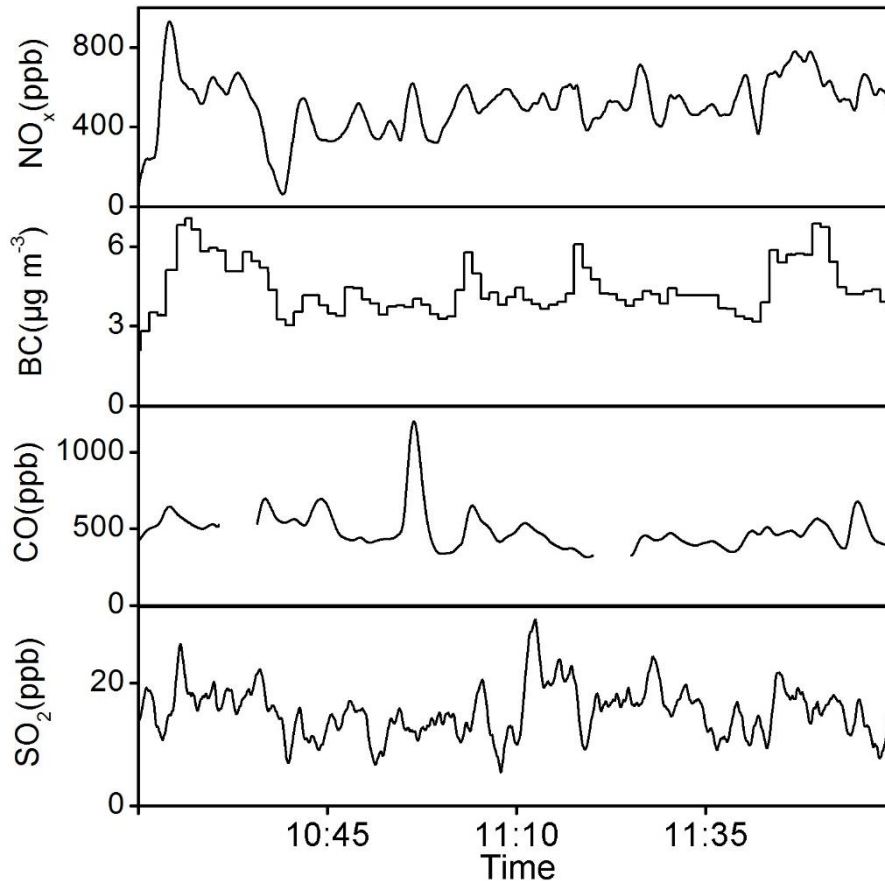


Fig. 2. The time series of the concentrations of NO_x , CO, SO_2 and BC measured on June 13, 2013.

3. Using the ratios of weighted vehicular emission factors to estimate the concentration enhancement of NO_x , CO, SO_2 and BC measured on highway

Based on the reported vehicular emission factors (Shen et al., 2015; Cai and Xie, 2007, 2010; Lei et al., 2011) and the vehicle composition (Chinese Automotive Technology & Research Centre, 2015) in Hebei province, where we conducted the most of the mobile measurements, we estimated the weighted vehicular emission factors on the highways (Table 2). The factors for NO_x , CO, SO_2 , and BC during our measurements were 2.9 g km^{-1} , 4.8 g km^{-1} , 0.04 g km^{-1} , and 0.01 g km^{-1} , respectively. If we assumed 400 ppb as the NO_x concentration enhancement on-road caused by vehicular emission, using the ratios of the weighted vehicular emission factors, the estimated enhancements of CO, SO_2 and BC concentrations on-road emitted by vehicles were 240 ppb, 3 ppb and $1 \text{ } \mu\text{g m}^{-3}$. These are at the similar levels of those enhancement showed in Table 1, suggesting vehicular emission is the main source for the enhancement of the on-road concentrations of NO_x , CO, SO_2 , and BC. However, the

enhancement of CO and SO₂ concentrations on highway were mostly less than 30%, this provides further evidence that CO and SO₂ concentrations on highway were dominated by regional background; and vehicular emission was not the main source.

Table 2. Estimated weighted vehicular emission factors of CO, NO_x, SO₂ and BC during mobile measurements in Hebei Province, China.

Vehicle type	Composition	Emission factors			
		CO ^[1] g km ⁻¹	NO _x ^[1] g km ⁻¹	SO ₂ ^[2] g km ⁻¹	BC ^[3] g km ⁻¹
Diesel bus	20%	1.2	11	-	-
Medium-duty diesel vehicles	20%	1.5	6.4	-	-
Light-duty gasoline vehicles	50%	4.4	1.3	-	-
Heavy-duty vehicles	10%	1.6	6.6	0.4	0.11
Weighted emission factor		2.9	4.8	0.04	0.01

^[1] Shen et al., 2015; ^[2] Cai and Xie, 2007; ^[3] Lei et al., 2011

In summary, both the comparison of on- and off-road measured concentrations and the vehicular emission factors provided the evidences:

- (1) Vehicular emission is the main source for the enhancement of the on-road concentrations of NO_x, CO, SO₂, and BC;
- (2) The high enhancement of NO_x concentration on highway suggesting NO_x on highway was mainly from vehicular emission;
- (3) CO and SO₂ concentration have up to 20% and 31% average enhancement on highway, suggesting that CO and SO₂ on highway were mainly from regional background.
- (4) The lifetimes of CO and SO₂ in the atmosphere are longer than that of NO_x, this is likely the main reason that the concentrations of CO and SO₂ measured on high way were dominated by regional background, while NO_x was not.

Changes in Manuscript: we have added detail discussions of the influence of traffic emission; please refer to the revised manuscript, from Page 15 Line 291 to Page 19 Line 391.

Comment No. 2. *Another concern is that the reported data may have major flaws. The extremely high NO_x values are not consistent with moderate values of the other three pollutants. If the NO_x 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 ~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.*

Response to Referee Comment No. 2:

The extremely high NO_x values have been reported in previous studies (Kolb et al., 2004; Westerdahl et al., 2005). As described in the response to the comment 1, we provided the evidences that the extremely high NO_x values were contributed by vehicular emission, while the moderate values of CO and SO₂ were mainly from regional background, plus about 20-30% from vehicular emission.

In previously reported studies of on-road mobile measurements, the daily concentration of NO_x could be higher than 400 ppb (Westerdahl et al., 2005). 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 NO_x and BC measured had similar trends (Fig. 2) and correlated each other relatively well (Fig. 3). Based on this regression, we can estimated the ratio of emission factors of BC and NO_x to be 0.004:1, which is close to the previously reported diesel vehicular emission factors in Beijing (0.008:1) (Wang et al., 2012). It suggests that diesel vehicular emissions had a large impact on the BC and NO_x concentrations measured on highways in NCP.

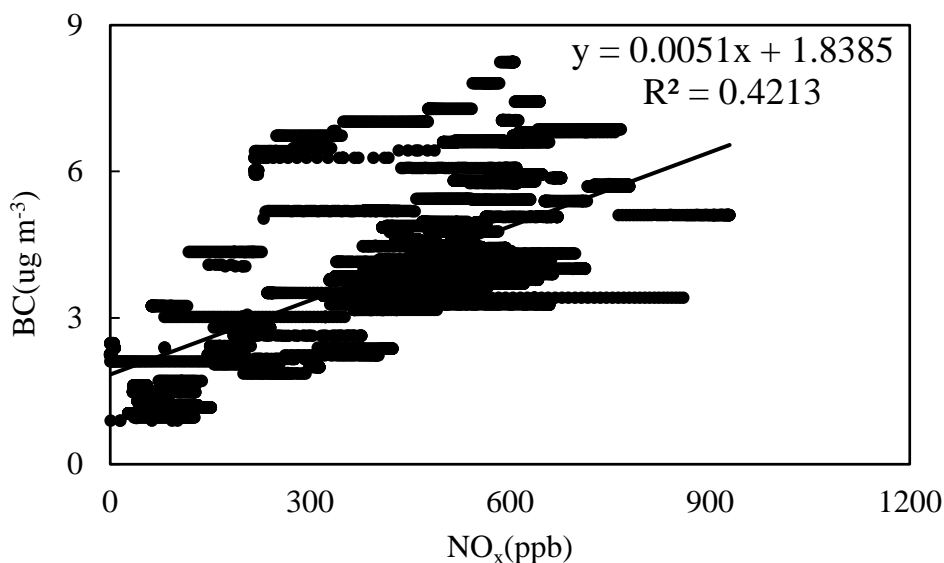


Figure 3. The regression of the concentrations of NO_x and BC measured on-road on in June 13, 2013.

Changes in Manuscript: we have added the discussion of the enhancements of CO and BC measured on road, please refer to the revised manuscript, from Page 15 Line 291 to Page 16 Line 324.

Comment No. 3. *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 to Referee Comment No. 3: We did make measurements off road to compare with the on-road data to check the impact of road vehicle emissions, please refer to our response to comment 1.

In previous study, we also did inter-comparison of our mobile measurement platform with a monitoring station in the campus of Peking University (Wang et al., 2009).

For this study, each time before an experiment, we did a calibration to obtain calibration curves, e. g. on June 16, 2013 (Fig. 4), and after the experiment we did another calibration and recorded the span drifts. The span drifts were less than 10%.

For example, according to the calibration on June 23, 2013, the span drifts of NO, SO₂ and CO were 29 (365, span) ppb, 9 (160) ppb, and 0.1 (7.4) ppm, respectively.

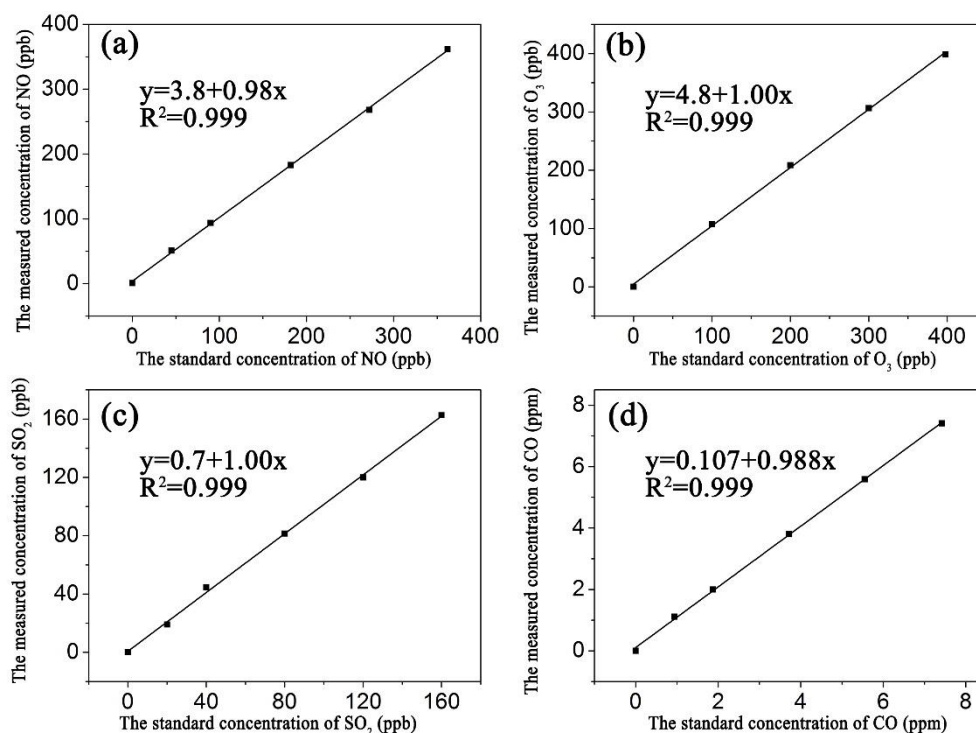


Figure 4. Calibration curves of NO, O₃, SO₂, and CO analyzers on June 16, 2013.

More in-depth analysis and discussion of the data set have been added in the revised version; please refer to Page 15 Line 291 to Page 19 Line 391.

Changes in Manuscript: we have added more detail description above the calibration method; please refer to the revised manuscript, from Page 7 Line 124 to Line 130. We also added in-depth discussion; please refer to Page 15 Line 291 to Page 19 Line 391 in the revised version.

Referee #2:

Comment No. 1. *Most seriously, I have concerns about the representativeness of the dataset, given that the measurements were only on expressways. Just because vehicle emissions aren't major sources in the regional inventory, they are very important*

when sampling in the aggregate exhausts of other vehicles. Therefore, arguments stating that they are small regionally do not hold for these measurements. Indeed, the NO_x values are argued as being influenced by vehicles – why wouldn't SO_2 , CO, and black carbon be impacted? CO has a two-month lifetime on average, yet measurements of CO on a highway are typically much higher than, say, at a park 1 km away. It is the enhancements above the regional background that matter, and trying to separate local (road) vs. regional background measurements is not clear without extensive, other data. Did the authors drive off-expressway to examine how their measurements changed when off-expressway and (ideally) away from other vehicles (parks)? How much traffic was around the mobile laboratory? Were higher concentrations reported while in traffic jams (surrounded by cars)? The extent to which the mobile vehicle measures “expressway” versus more “regional background” levels will vary depending on the traffic density, winds, and stability. Consider the two extreme cases: 1) on a road with no other vehicles, the mobile lab will measure the regional background; 2) while stuck in traffic in a multi-lane city expressway, the measurements are clearly just vehicle emissions. How this measurement footprint changes versus location/day/traffic/wind is difficult to quantify without more measurements (see comment 3 below for more details). WFR-CHEM or FLEXPART can't model at roadway scales, of course, so there has to be some criteria applied to assess such influences. On p. 7, it is stated that traffic jams were avoided – this is rather ambiguous. How was this defined? In all this driving, there was never a traffic jam?

Response to Reviewer Comment No. 1: We agree that we need to provide more evidences to justify the representativeness of our dataset for the regional background concentrations of air pollutants. Following the comments and suggestions of the reviewer, we have rewritten the manuscript and provided more evidences.

1. Comparing the on- and off-road measurements of air pollutants to estimate the enhancement of air pollution on highway above the regional background

Each on-road measurement trip started from a parking lot in a highway service center and ended at the parking lot of another service center. The parking lots are about 150 m away from highway, such as those in service centers Dezhou (DZ) and Xizhaotong (XZT) (Fig. 1). Using the difference of the concentrations of air pollutants measured in a parking lot and on highway, we can estimate the level of the enhancement of the concentrations of air pollutants on highway above regional backgrounds.

Table 1 shows the 5 min averaged concentrations of NO_x, CO, SO₂, and BC measured in parking lots and on highways, and the difference between the on- and off-road concentrations. The concentrations in parking lots were measured for 5 mins before driving or after parking with engine turned off, and the concentrations on highways were measured for 5 mins after entering highways or 5 mins before entering the service centers.

The concentrations of NO_x measured on highway show drastic enhancement than those measured off-road, from 19 to 449 ppb, or 43% to 1658% (510±61%, mean± Standard deviation), while other pollutants have much lower enhancement or even reduction in concentrations. The difference between the on-road and off-road concentrations of CO ranged from -478 to 145 ppb, or -28% to 34% (7±22%); for SO₂, it is -26 to 13 ppb or -18% to 175% (52±59%); for BC, it is -0.7 to 4.7 μg m⁻³ or -11% to 261% (85±90%).

The 175% enhancement of SO₂ on July 12 could be due to the much lower SO₂ concentration, 1.2 to 3.3 ppb, than those measured in other days. The rain on July 12 is likely the major reason for the much lower concentrations of SO₂ (3 ppb) and NO_x (63 ppb), while CO and BC, which are much less water soluble, show no significant difference than those measured in other days. If we ignore the results of SO₂ and NO_x on July 12, then the difference between the on- and off-road concentrations were 82% to 1658% (510±61%) for NO_x and were -18% to 87% (31±31%) for SO₂.

Apparently, vehicular emission is the major source lead to the 82% to 1658% enhancement of NO_x concentrations on highway. The enhancement of CO and SO₂ concentrations on highway were mostly less than 30%, suggesting vehicular emission is not the main source for CO and SO₂ on highway, regional background is the dominant factor determine their concentrations.

2. Time series of the concentrations of air pollutants measured on highway

Figure 2 shows the concentrations of NO_x, CO, SO₂, and BC measured on highway on a typical day, June 13, 2013. Apparently, the concentration of BC follows the trend of NO_x concentration and those of CO and SO₂ did not. This suggests the high enhancement of NO_x and BC concentration on highway was due to the vehicular emission, while the concentrations of CO, SO₂ were not.

3. Using the ratios of weighted vehicular emission factors to estimate the concentration enhancement of NO_x, CO, SO₂ and BC measured on highway

Based on the reported vehicular emission factors (Shen et al., 2015; Cai and Xie, 2007, 2010; Lei et al., 2011) and the vehicle composition (Chinese Automotive Technology & Research Centre, 2015) in Hebei province, where we conducted the most of the mobile measurements, we estimated the weighted vehicular emission factors on the highways (Table 2). The factors for NO_x, CO, SO₂, and BC during our measurements were 2.9 g km⁻¹, 4.8 g km⁻¹, 0.04 g km⁻¹, and 0.01 g km⁻¹, respectively. If we assumed 400 ppb as the NO_x concentration enhancement on-road caused by vehicular emission, using the ratios of the weighted vehicular emission factors, the estimated enhancements of CO, SO₂ and BC concentrations on-road emitted by vehicles were 240 ppb, 3 ppb and 1 μg m⁻³. These are at the similar levels of those enhancement showed in Table 1, suggesting vehicular emission is the main source for the enhancement of the on-road concentrations of NO_x, CO, SO₂, and BC. However, the enhancement of CO and SO₂ concentrations on highway were mostly less than 30%, this provides further evidence that CO and SO₂ concentrations on highway were dominated by regional background; and vehicular emission was not the main source.

In summary, both the comparison of on- and off-road measured concentrations and the vehicular emission factors provided the evidences:

- (1) Vehicular emission is the main source for the enhancement of the on-road concentrations of NO_x, CO, SO₂, and BC;
- (2) The high enhancement of NO_x concentration on highway suggesting NO_x on highway was mainly from vehicular emission;
- (3) CO and SO₂ concentration have up to 20% and 31% average enhancement on highway, suggesting that CO and SO₂ on highway were mainly from regional background.
- (4) The lifetimes of CO and SO₂ in the atmosphere are longer than that of NO_x, this is likely the main reason that the concentrations of CO and SO₂ measured on high way were dominated by regional background, while NO_x was not.

4. Possible traffic jams and high concentrations of air pollutants

Each on-road measurement trip started from a parking lot in a highway service center and ended at the parking lot of another service center. The driven speed of the mobile platform was never lower than 80 km h⁻¹ on highways, this is one of the evidences that we encountered no traffic jams, and the number of vehicles around our mobile platform seldom increased.

During our measurements, as shown in Fig. 3 in the manuscript, we always measured high concentrations of air pollutants near large cities. As the time series of the concentrations of NO_x, CO, SO₂ and BC measured on June 13 (Fig. 5) show, when approaching Baoding, CO concentration gradually increased from 500 ppb at 50 km away from Baoding to 1200 ppb at 23 km away from Baoding. The same is SO₂ (from 10 ppb to 40 ppb) and BC (from 4 μg m⁻³ to 8 μg m⁻³), while NO_x does not show the same trend. When at 15 km away from Baoding, we see another increase of all the pollutants, and the same decrease trend after at 10 km from Baoding. The same continuously increasing trends of SO₂ and CO, started at 50 km to 15 km away from Baoding, indicated the regional transport of SO₂ and CO. Started at 15 km, away from Baoding, all the four pollutants had a peak with about 10 km width, which indicated the urban emissions.

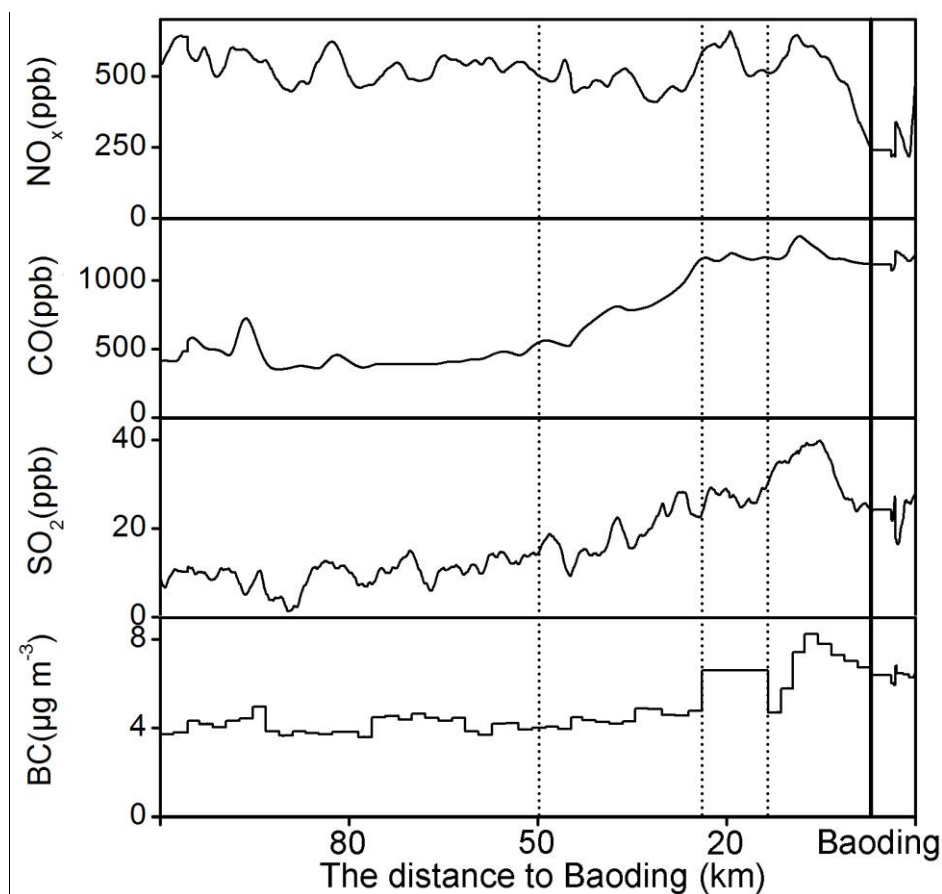


Fig. 5. The time series of the concentrations of NO_x, CO, SO₂ and BC measured on June 13, 2013 (the three dashed lines marked the 50 km, 23 km, 15 km away from Baoding, and the solid line is at Baoding).

In conclusion, in this experiment, vehicular emissions contributed less than 30% of CO and SO₂ measured on-road, we can use the CO and SO₂ measurement to study the spatial distributions of CO and SO₂ in NCP. Moreover, the hot spots measured near large cities were mainly from local emissions.

Changes in Manuscript: we have added more detailed discussions of the influence of traffic emission; please refer to the revised manuscript, from Page 15 Line 291 to Page 19 Line 391.

Comment No. 2. *A large number of values are reported, and too often this reads as a “data dump” with little insight besides vague generalizations. Tables comparing select case studies may be helpful, particularly highlighting the different spatiotemporal domains of the comparisons.*

Response to Reviewer Comment No. 2: Accept.

Changes in Manuscript: we have added a table (Table 3) in the manuscript to summarize the results and highlighted the different spatiotemporal domains of the comparisons in the revised paper. Please refer to the revised manuscript:

Page 21, Line 416, ...on July 2 (route 1) and July 7 (route 5), respectively.

Page 21, Line 422, were regions with low emissions (Fig. 3)...

Page 21, Line 431, On June 24 (route 1), June 14 (route 4) and June 15 (route 5),...

Page 22, Line 443, ..., as the same route on June 14 (Fig. 7),...

Page 24, Line 479, ...different wind fields. Route 2 experienced southwest winds on June 12 and July 3,...

Page 24, Line 483, ...the wind direction on route 2 in the central NCP,...

Page 24, Line 498, ...the wind fields were similar on route 3 on June 13 and July 4 (Fig. 9),...

Table 3. Trip-average concentrations of SO₂, CO and BC and wind directions on those days discussed in the manuscript

Route	Date	Concentrations (mean ± SD, ppb)			Wind directions
		SO ₂	CO	BC	
Route 1	July 2	4.5±2.3	550±240	5.0±2.6	northwest
Route 5	July 7	7.0±3.0	1090±320	6.5±2.7	east
Route 1	June 24	15±5.8	1300±330	8.0±1.4	south
Route 4	June 14	26±7.9	1200±230	6.5±1.5	south
Route 5	June 15	28±7.1	1600±370	7.0±1.9	south
Route 4	July 6	14±7.6	1100±450	4.6±1.8	south
Route 2	June 12	26±11	950±440	4.8±2.2	southwest
Route 2	June 18	27±10	1030±530	6.8±2.3	northwest
Route 2	June 25	27±16	1020±680	6.5±3.3	stable
Route 2	July 3	23±15	989±450	5.7±2.6	southwest

Comment No. 3. *Some comparisons to ground-based sites were noted (also to literature), but even these were rather trivial and limited. Comparing a transect along a nearby road to a nearby stationary site (e.g. “YC” in text), it is unclear if the mobile laboratory stopped there for a certain length of time or if it just passed by the site as part of the drive. How close to the site was the vehicle located and for what time domain for the comparisons listed (e.g. Fig. 2, p15, lines 300-310, etc.)? By knowing the mean wind, one should be able to link space-time scales at some level. Instead, just a date is given – did the authors integrate for the entire day for both measurements? Given the contamination variable contribution of vehicle exhaust in their data set, quantifying agreement is with uncertainties.*

Response to Reviewer Comment No. 3: Accept. We have added more information about the locations of these stationary sites in the manuscript:

1. The straight-line distance from these stationary sites to the nearest measuring roads: GC station to route 1, 3 km; QZ station to route 2, 54 km; YC station to route 2, 5 km.
2. The time domain for the comparisons: These stationary data were hour-average, and the concentrations plotted in Fig. 2 in the manuscript were hour-average results when our mobile laboratory passed by these stations.

Furthermore, the footprint maps (Fig. 5 in the manuscript and Fig. S4 in the supplement) show the wind directions and wind speeds at these sites.

Changes in Manuscript: we have added more information about the locations of these stationary sites, please refer to the revised manuscript, from Page 10 Line 204 to Page 11 Line 210.

Comment No. 4. *As the manuscript noted, concentrations are impacted by wind direction (source regions upstream), local sources (which are never really specified – just stating “urban” area), location (e.g. in the central region wind direction plays less of an influence than at the margins) and boundary layer height (e.g. p. 21, line 421-425). It is hard to interpret the variations observed in Fig. 2 – which are caused by which of the above factors and which ones dominate (and when)? Reasonable examples are provided in the following text of all of the above, but in the end, I have difficulty interpreting the importance of each of these processes in the full dataset.*

Response to Reviewer Comment No. 4: Agree. We discussed these four impactors in different sections in the manuscript, and to make it clearer, we added following sentences and paragraphs in the manuscript:

“Precipitation caused the low concentrations of NO_x and SO₂ in the last experiment in NCP. And all measuring areas were affected by heavy rain, so the concentrations of NO_x and SO₂ on all routes were low.”

“We found all air pollutants measured near large cities were at high level. According to the analysis in section 3.2, they were mainly from local emissions of cities. Also, according to the estimated emission inventories, these cities were major sources of air pollution which caused the concentrations of air pollution high around them.”

Also, we added a conclusion paragraph at the end of section 3.3:

“We discussed five impactors: local emission, precipitation, location, wind direction and boundary layer height. The influence of local emission reflected in the spatial distribution of concentrations (Fig. 3 in the manuscript). Hot spots were found near cities. However, for route-average results (Fig. 2 in the manuscript), local emission plays a minor role in the distribution of concentrations, because the routes were mainly in suburb. The large reduction in SO₂ and NO_x concentration measured on July

12 along all routes were caused by precipitation. The persistent high concentrations of pollutants in route 2 were associated with the location of it, which was surrounded by high emission areas. Besides the route 2, the different concentrations in one route in different measurements were mainly from different wind directions. And we also found one case that the concentrations changed a lot between June 13 and July 4 in route 3 under the similar wind directions. Based on the model results, boundary layer height might explain the change.”

Changes in Manuscript: we have added discussion of the five impactors, please refer to the revised manuscript, from Page 14 Line 278 to Line 280, from Page 20 Line 395 to Line 399, and from Page 26 Line 537 to Line 549.

Comment No. 5. *Experimental methods: what are the precisions and uncertainties in the measurements (e.g. NO_x precision; CO precision and accuracy; SO₂ precision and accuracy; BC same thing), only NO_x had an uncertainty noted (but no precision). Did the laboratory studies/calibrations done on non-driving days agree or disagree with stated manufacture specifications?*

Response to Reviewer Comment No. 5: Accepted. We added the precisions and uncertainties of these analyzers in section 2.1 in the manuscript (Table 4). The NO_x precision was 1% and the uncertainty was less than 10%. The CO precision was 1%, and the uncertainty was less than 100 ppb. The SO₂ precision was 0.5%, and the uncertainty was less than 10%. The BC precision was 0.1 μg m⁻³ and the uncertainty was less than 1%. These precisions were from manufacture specifications and uncertainties were from calibrations. Each time before an experiment, we did a calibration to obtain calibration curves, e.g. on June 16, 2013 (Fig. 4), and after the experiment we did another calibration and recorded the span drifts. The span drifts were less than 10%. For example, according to the calibration on June 23, the span drifts of NO, SO₂ and CO were 29 (365, span) ppb, 9 (160) ppb and 0.1 (7.4) ppm. We added this information in the manuscript.

Table 4. Precisions and uncertainties of air pollutants analyzers used in our experiments

	NO _x	CO	SO ₂	BC
Precision	1%	1%	0.5%	0.1 μg m ⁻³
Uncertainty	<10%	<100 ppb	<10%	<1%

Time resolution	30 s	40 s	120 s	6 s
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Changes in Manuscript: we have added more detailed information of the precisions and uncertainties of these analyzers, please refer to the revised manuscript, from Page 6 Line 115 to Line 130.

Comment No. 6. *To what extent was the data coverage for the 5 campaigns? 100%? Rain is mentioned on p. 8, line 152 – what about other data dropouts (if they existed)?*

Response to Reviewer Comment No. 6: Accept. The major reasons for data missing were the computer crashing and rain. Rain caused the missing data on route 3, route 4 and route 5 in experiment 3, and on route 5 in experiment 5. Computer crashing caused the missing data on route 4 and route 5 in experiment 2 (Table 5). Also, the computer crashing caused data missing during every trip (Table 6). We added the information in the manuscript.

Table 5. Measurement data and routes in the 5 campaigns

Date	Route 1	Route 2	Route 3	Route 4	Route 5
June 11–June 15	June 11	June 12	June 13	June 14	June 15
June 17–June 20	June 17	June 18	June 20	-	-
June 24–June 25	June 24	June 25	-	-	-
July 2–July 7	July 2	July 3	July 4	July 6	July 7
July 11–July 15	July 11	July 12	July 14	July 15	-

Table 6. Data coverage of every trip

Date	Start time	End time	Data coverage	Reason for data missing
June 11	10:00	14:00	67%	Computer crashing
June 12	10:23	14:23	92%	Computer crashing
June 13	10:15	14:00	100%	
June 14	9:55	13:55	100%	
June 15	9:58	14:14	99%	Computer crashing
June 17	10:40	14:20	78%	Computer crashing
June 18	9:55	13:35	80%	Computer crashing
June 20	10:15	13:05	85%	Computer crashing
June 24	10:50	14:10	66%	Computer crashing
June 25	10:20	14:20	83%	Computer crashing

July 2	10:23	14:15	72%	Computer crashing
July 3	10:27	13:35	72%	Computer crashing
July 4	10:10	11:46	100%	
July 6	9:58	14:22	98%	Computer crashing
July 7	10:12	14:46	74%	Computer crashing
July 11	10:22	14:30	66%	Computer crashing
July 12	10:10	14:07	84%	Computer crashing
July 14	10:13	11:42	100%	
July 15	10:12	15:05	88%	Computer crashing

Changes in Manuscript: we have added information of data missing, please refer to the revised manuscript, from Page 8 Line 160 to Line 164, and the supplement Table S1 and Table S2.

Comment No. 7. *p. 12, “low levels of SO₂”: 10 ppbv SO₂ is not low, perhaps compared to the past in China. But I’m not sure how this mobile lab study over a very limited time/space can verify that SO₂ has decreased and confirmed policy desulphurization devices. No comparison to past SO₂ measurements in these locations is noted, so this seems unsubstantiated from this study.*

Response to Reviewer Comment No. 7: Accept.

Changes in Manuscript: we deleted the “low levels of SO₂” and related discussions in the manuscript. Please refer to the revised manuscript, from Page 13 Line 254 to Line 255.

Comment No. 8. *Some data should be made available to the community on a website, not “on request”, given the political sensitivity of Chinese air pollution data. What metadata will be provided, e.g. GPS, vehicle information (speed), met data, etc.?*

Response to Reviewer Comment No. 8: Agree. We are happy to provide our data on a website. We had a website for this purpose during CAREBEIJING Campaign. However, this site is closed due to lack of fund. We are working on it and hoping to reinstall the web site in this year. We will provide data of GPS, vehicle speed, meteorology, concentrations of air pollutants.

Changes in Manuscript: we have added more information about data availability; please refer to the revised manuscript, from Page 28 Line 570 to Line 571.

Comment No. 9. *I like the FLEXPART trajectory attribution, but again this is a qualitative comparison as the upwind values were not measured. So while it makes sense that northerly winds off the mountains will decrease values nearby, I don't think this was particularly profound.*

Response to Reviewer Comment No. 9: Agree. This argument was based on a general situation that the air quality in the north border of the North China Plain was clean, because the emissions of air pollutants were low in that area (Fig. 3 in the manuscript). To support this argument, we collected the data of air pollutants concentrations measured on the Huairou site (116.63 °E, 40.32 °N) on July 2, 2013 from State Control Monitoring Sites database (<http://www.mep.gov.cn/hjzl/>). The average concentrations of CO, NO₂, and SO₂ from 9:00 to 15:00 on that day were 0.3 ppm, 3.4 ppb, and 3.3 ppb, respectively. It indicates that the concentrations of air pollutants in the north border of NCP were low.

Changes in Manuscript: we have added explanation of the clear air from north of NCP, please refer to the revised manuscript, from Page 23 Line 458 to Line 460.

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1 **Distribution and Sources of Air pollutants in the North**
2 **China Plain Based on On-Road Mobile Measurements**

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17

18 **Abstract.** The North China Plain (NCP) has been experiencing severe air pollution
19 problems with rapid economic growth and urbanisation. Many field and model studies
20 have examined the distribution of air pollutants in the NCP, but convincing results
21 have not been achieved mainly due to a lack of direct measurements of pollutants over
22 large areas. Here, we employed a mobile laboratory to observe the main air pollutants
23 in a large part of the NCP from June 11 to July 15, 2013. High median concentrations
24 of sulphur dioxide (SO₂) (12 ppb), nitrogen oxides (NO_x) (NO+NO₂; 452 ppb), carbon
25 monoxide (CO) (956 ppb), black carbon (BC; 5.5 μg m⁻³) and ultrafine particles
26 (28350 cm⁻³) were measured. Most of the high values, i.e., 95 percentile
27 concentrations, were distributed near large cities, suggesting the influence of local
28 emissions. In addition, we analysed the regional transport of SO₂ and CO, relatively
29 long-lived pollutants, based on our mobile observations together with wind field and
30 satellite data analyses. Our results suggested that, for border areas of the NCP, wind
31 from outside would have a diluting effect on pollutants, while south winds would
32 bring in pollutants accumulated during transport through other parts of the NCP. For
33 the central NCP, the concentrations of pollutants were likely to remain at high levels,
34 partly due to the influence of regional transport by prevalent south–north winds over
35 the NCP and partly by local emissions.

36 **Keywords:** North China Plain, Air pollution, Distribution, On-road mobile
37 measurements

38

39 **1. Introduction**

40 The North China Plain (NCP) is a geographically flat region in the northern part of
41 Eastern China, which includes Beijing, Tianjin, most of Hebei, Henan and Shandong
42 provinces, and the northern parts of Anhui and Jiangsu provinces. This region is
43 surrounded by the Yan Mountains to the north, the Taihang Mountains to the west and
44 the Bohai Sea to the east. The NCP covers an area of 300,000 km², which corresponds
45 to about 1/32 of the total area of China, but is home to approximately 1/5 of the
46 Chinese population. The NCP is the political, economic and agricultural centre of
47 China. Along with rapid economic growth and urbanisation, the NCP has been
48 experiencing severe air pollution problems (Donkelaar et al., 2010). On a global scale,
49 the NCP is a hotspot of nitrogen dioxide (NO₂), carbon monoxide (CO), sulphate and
50 particulate matter (PM) concentrations, according to both satellite observations and
51 model simulations (Chin et al., 1996; Yu et al., 2010; Bechle et al., 2011; Streets et al.,
52 2013; Bucselo et al., 2013). The concentrations of PM with an aerodynamic diameter
53 $\leq 2.5 \mu\text{m}$ (PM_{2.5}) and PM₁₀ in the NCP are much higher compared to other rapidly
54 developing areas in China, such as the Yangtze River Delta (Hu et al., 2014).
55 According to the air-quality report published by the Chinese Ministry of
56 Environmental Protection, in 2013, 9 of the 10 most polluted cities in China were
57 located in the NCP. Severe pollution events occur frequently in this area. Therefore,
58 studies of air pollution problems in the NCP are essential to obtain general insights
59 into the unique patterns of air pollution in this area and for management of emissions
60 control policies by the government.

61 Over the past decade, there have been a number of investigations of air pollution in
62 the NCP taking advantage of observation sites, aircraft measurement platforms,
63 mobile laboratories, satellite data and air quality models. In the NCP, a network of
64 observation sites has been built for air pollution research, mostly located in and
65 around large cities, particularly Beijing (Xu et al., 2011; Xu et al., 2014; Wang et al.,
66 2013; Meng et al., 2009; Shen et al., 2011; Lin et al., 2011). Variability, sources,
67 meteorological and chemical impacts of air pollutants have been discussed by
68 analysing these observational results. The concentrations of long-lived pollutants have
69 been shown to be significantly influenced by wind, particularly the south and north
70 winds, indicating that regional transport plays an important role in urban air pollution.
71 In addition, model studies have yielded similar results in various areas of the NCP
72 (An et al., 2007; Zhang et al., 2008; Liu et al., 2013). Satellite data have indicated that
73 regional transport has a significant impact on the haze period in the NCP (Wang et al.,
74 2014). Thus, it is necessary to understand regional transport to address air pollution
75 problems in the NCP, which will require data on the distribution of air pollutants in
76 this region.

77 However, observational data from a single or limited number of measurement sites
78 cannot present the whole picture of air pollution in the NCP. A number of mobile
79 laboratory measurements (Johansson et al. 2008; Li et al., 2009; Wang et al., 2009;
80 Wang et al., 2011) and aircraft measurements (Huang et al. 2010; Zhang et al. 2011;
81 Zhang et al. 2009; Zhang et al. 2014) have been used to determine pollution
82 distributions mainly within the megacity of Beijing. There have been several reports

83 of model and satellite studies on the air pollution distribution in the NCP, or even the
84 whole of China (Wei et al., 2011; Zhao et al., 2013; Ying et al., 2014; Ding et al., 2015;
85 Ding et al., 2009). However, there are disagreements between these results, e.g.,
86 regarding the distributions of NO₂ in several hotspot areas produced from model
87 (CMAQ) and satellite (SCIAMACHY) measurements (Shi et al., 2008). Uncertainties
88 in model simulations such as emissions inventories, and in satellite measurements
89 such as the influence of clouds, can only be evaluated by measuring the spatial
90 distributions of pollutants over a large geographical area, which are still lacking.

91 In this study, we measured the concentrations of nitrogen oxides (NO_x), CO,
92 sulphur dioxide (SO₂), ultrafine particles and BC with a mobile laboratory platform in
93 the NCP. Satellite data and field wind measurements during the observation period
94 were also used. Our specific objectives were to collect a dataset showing the spatial
95 distribution of air pollutants in the NCP, and to characterise the regional transport
96 within and outside the NCP. This study was performed as part of the Campaigns of
97 Air Pollution Research in Megacity Beijing and North China Plain
98 (CAREBeijing-NCP 2013), and involved comprehensive stereoscopic observations,
99 including observations of two super sites, several routine sites, mobile laboratories
100 and model work. This paper focuses on the distribution and transport of pollutants in
101 the NCP, mainly based on data collected from a mobile platform.

102 **2. Experimental methods**

103 **2.1 Mobile laboratory and study area**

104 A mobile laboratory was built by our research group, details of which were previously
105 described (Wang et al., 2009). Briefly, this mobile laboratory was constructed in 2006
106 on an IVECO Turin V diesel vehicle (L = 6.6 m, W = 2.4 m, H = 2.8 m; payload = 2.7
107 metric tonnes). Instrumentation was powered by two sets of uninterruptible power
108 systems (UPS), consisting of three series of 48 V/110 Ah lithium batteries, which
109 could support all of the equipment operations without interruption for up to 5 h. The
110 inlet systems for our mobile laboratories were specifically configured to
111 accommodate the type of measurement requirements and the instrument suite to be
112 employed in specific field campaigns.

113 Instruments deployed on the mobile laboratory included those for studying NO_x,
114 CO, SO₂, BC and ultrafine particles. NO_x was measured using an NO_x analyser with
115 an Mo-converter (Ecotech model 9841A; Ecotech, Knoxfield, Melbourne, Australia).
116 CO was measured with a CO analyser by light absorption (Ecotech model 9830A).
117 SO₂ was measured using an SO₂ analyser with a fluorescence cell (Ecotech model
118 9850A). BC was measured using a multi-angle absorption photometer (MAAP;
119 Thermo model 5012; Thermo Scientific, Waltham, MA). The precisions, uncertainties
120 and time resolutions of these analysers were shown in table 1. The online
121 measurement data from these instruments were recorded with an industrial personal
122 computer.

Table 1. here.

Each time before an experiment, we did a calibration to obtain calibration curves, e.g. on June 16 in 2013 (Fig. S2), and after the experiment we did another calibration and recorded the span drifts. The span drifts were less than 10%. For example, according to the calibration on June 23 in 2013, the span drifts of NO, SO₂ and CO were 29 (365) ppb, 9 (160) ppb and 0.1 (7.4) ppm. In previous study we also did inter-comparison with monitoring station in the campus of Peking University (Wang et al., 2009).

Ultrafine particles were measured with a fast mobility particle sizer (FMPS, TSI 3090; TSI, Shoreview, MN), which covers particle sizes from 5.6 nm to 560 nm in 32 channels with a time resolution of 0.1 s. The data were recorded on a dedicated computer. Other auxiliary data including temperature, relative humidity, barometric pressure and GPS coordinates were also measured. The driving speed was kept stable at around 100 ± 5 km h⁻¹ to cover as much distance as possible with the 5 h of power supplied by the lithium batteries.

To establish the spatial distribution and characterise the regional transport of air pollutants in the NCP, the routes for the mobile measurements were specially designed to cover important emissions hotspots (Fig. 1) and to map large areas of the NCP. The routes included the municipalities of Beijing and Tianjin, most of Hebei province, and part of Shandong province, which is about 300 km wide from the west to the east and 400 km long from the north to the south, covering most of the NCP. To avoid traffic

144 jams and rough roads, only expressways were chosen for all routes. Limited by the
145 duration of battery power and the variability of boundary layer height, we could not
146 cover all routes in one trip. Instead, we divided the routes into five parts. Route 1 was
147 along the Taihang Mountains from Beijing to Shijiazhuang, located in the western part
148 of the NCP. Routes 2 and 3 were from Shijiazhuang to Dezhou and Cangzhou to
149 Baoding, respectively, which were generally located in central NCP. Routes 4 and 5
150 were from Tianjin to Beijing and around the south of Beijing, located in northern NCP.
151 In addition, we ran each route in one day. Two days were also needed for calibration
152 and maintenance of instruments. Therefore, it took one whole week to conduct a
153 single experiment. In total, six experiments, including one pre-test study, were
154 designed from June 1 to July 15, 2013. The pre-test study was conducted between
155 June 1 and June 7, and five formal repeated experiments were conducted between
156 June 11 and July 15 (Experiment 1 [E1], June 11–June 15; E2, June 17, June 18 and
157 June 20; E3, June 24–June 25; E4, July 2–July 7; E5, July 11–July 15). All trips were
158 started at about 09:00 and ended at about 14:00 to ensure that the boundary layer was
159 relatively stable during the observation period in 1 day.

160 The major reasons for data lacking were the computer crashing and rain. Rain
161 caused the missing data on route 3, route 4 and route 5 in experiment 3, and on route 5
162 in experiment 5. Computer crashing caused the missing data on route 4 and route 5 in
163 experiment 2 (Table S1). Also, the computer crashing caused data missing during
164 every trip (Table S2)

Figure 1 here.

2.2 The trajectories model

A Lagrangian particle dispersion model, FLEXPART-WRF version 3.1 (Brioude et al., 2013; Stohl et al., 1998; Stohl et al., 2005; Fast and Easter, 2006), was used to determine the origin and transport pathways of the air mass arriving at the vehicle-based mobile measurement laboratory. The wind field used to drive FLEXPART was the time-averaged wind provided by the WRF with temporal intervals of 10 min and horizontally spatial resolution of 2 km (The details of the mesoscale meteorological model is described in S2.1). FLEXPART simulates the transport and dispersion of tracers by calculating the backward trajectories of multitudinous particles, which are termed plume back trajectories. In this model, turbulence in the planetary boundary layer (PBL) is parameterised by solving the Langevin equation, and convection is parameterised using the Zivkovic Rothman scheme (Stohl et al., 2005). To improve the accuracy of the trajectory calculation, we used high-resolution WRF simulation domain 4 outputs as the input meteorological conditions for the FLEXPART model. The turbulence, convection and boundary layer height were computed along the trajectories of tracer particles using the WRF output data. Backward integration was performed every 5 min during the mobile observation period in June 2013. For each integration, 2000 stochastic particles were released initially from within a box $1 \times 1 \text{ km}^2$ in horizontal extent and 1–50 m vertical height above ground centred on the mobile measurement laboratory. A total of 2000 inert

186 tracer particles were released about every 5 min along the route of the vehicle. For
187 each release, the backward trajectories were simulated for at least 12 h, and the
188 particle locations were output every 10 min for analysis. The 12 h length of the
189 backward trajectories was chosen as a trade-off to adequately sample the history of
190 the air masses over the region of interest, while decreasing the trajectory error (Stohl,
191 1998; Zhang et al., 2012).

192 The footprints of backward trajectories were calculated to present plume
193 trajectories. Footprints in this context refer to the total residence times of released
194 particles, which were calculated following Ashbaugh et al. (1985) and de Foy et al.
195 (2009) by counting the accumulated number of particles during the integration within
196 each cell of a 2×2 km² grid. Various transport and diffusion patterns can well be
197 described by these footprints analyses (Zhang et al., 2012).

198 **2.3 Stationary measurement sites and the fire data**

199 Concentrations of air pollutants, including NO_x, SO₂, CO and BC, were measured
200 simultaneously at three stationary measurement sites during CAREBeijing-NCP 2013.
201 These were rural sites located at Gucheng, Hebei province (GC, 39.13°N, 115.67°E),
202 Quzhou, Hebei province (QZ, 36.78°N, 114.92°E) and Yucheng, Shandong province
203 (YC, 36.67°N, 116. 37°E) (Fig. 1). The GC stationary site was near route 1, and QZ
204 and YC stationary sites were near route 2. The straight-line distances from these
205 stationary sites to the nearest measuring roads were 3 km from GC station to route 1,
206 54 km from QZ station and 5 km from YC station to route 2. And these stationary data

207 were hour-average. The concentrations plotted in Fig. 2 were hour-average results
208 when our mobile laboratory passed by these stations. Furthermore, the footprint maps
209 (Fig. 7 and Fig. S6) show the wind directions and wind speeds at these sites.

210 The main pollutants at these sites were measured using commercial instruments. At
211 the QZ site, gas analysers were used to measure NO_x (Ecotech model 9841A), CO
212 (Ecotech model 9830A) and SO₂ (Ecotech model 9850A). At GC and YC stations, gas
213 analysers were used to measure NO_x (Thermo model 42C), CO (Thermo model 48i)
214 and SO₂ (Thermo model 48i), and BC was measured by MAAP (Thermo model
215 5012).

216 Fire data were obtained from the Moderate Resolution Imaging Spectroradiometer
217 (MODIS) installed in Terra and Aura. The territory passing times were 10:30 (local
218 time) and 13:30 (local time) for Terra and Aura, respectively. Fire images were
219 obtained from EOSDIS Worldview (NASA, <https://earthdata.nasa.gov/labs/worldview>
220).

221 **3. Result and discussion**

222 **3.1 Distribution of air pollutants**

223 BC, NO_x, CO and SO₂ were measured on five routes during the experiment to
224 determine the concentrations of air pollutants on the routes and their spatial
225 distributions in the NCP. Figure 2 shows the results of our mobile measurements
226 obtained in 19 trips on the five routes from June 11 to July 15. The mean and median

227 concentrations of BC, NO_x, CO and SO₂ were 5.8 and 5.5 μg m⁻³, 422 and 452 ppb,
228 1006 and 956 ppb and 15 and 12 ppb, respectively, in the whole study. These high
229 values were consistent with previous measurements of most pollutants at stationary
230 measurement sites in the NCP except for NO_x. For example, the measured
231 concentrations of NO_x, SO₂ and CO were 62.7 ± 4.0 ppb, 31.9 ± 2.0 ppb and 1990 ±
232 130 ppb in an urban site in the courtyard of China Meteorological Administration in
233 the Beijing area from November 17, 2007, to March 15, 2008 (Lin et al., 2011). These
234 values were 13–50 ppb, 5.7–30.3 ppb and 1100–1800 ppb at an urban site in Wuqing
235 (between Beijing and Tianjin) from July 9, 2009, to January 21, 2010 (Wu et al.,
236 2011); and 28.4 ppb, 17.2 ppb and 1520 ppb at the GC site from July 2006 to
237 September 2007 (Lin et al., 2009). In addition, the concentration of NO₂ measured at
238 the YC site from June 18 to June 30 was about 20 ppb (Wen et al., 2015). This
239 comparison with stationary site measurements suggested that our mobile
240 measurements reflected the heavily polluted conditions in the NCP, which ensured its
241 feasibility in profiling the distributions of these air pollutants.

242 Figure 2 here

243 The levels of CO, NO_x and BC here were also comparable to those in previous
244 mobile laboratory measurements in European and American cities. Bukowiecki et al.
245 (2002) measured CO in Zürich, Switzerland, and the average concentration was about
246 600 ppb. Hagemann et al. (2014) measured NO_x in Karlsruhe in Germany, and the
247 average concentration was about 20 ppb. In the USA, NO_x was around 50 ppb in

248 Somerville (Padró-Martínez 2012), 200 ppb during rush hour in Boston (Kolb et al.,
249 2004) and ranged from 230 ppb to 470 ppb in Los Angeles (Westerdahl et al., 2005).
250 Padró-Martínez et al. (2012) also measured BC in Somerville, and reported average
251 concentrations of about $1 \mu\text{g m}^{-3}$. As these measurements were obtained in
252 heavy-traffic areas in large cities, and our results were measured over a large region,
253 we concluded that the air pollution problems in the NCP are among the worst in the
254 world. In contrast to these pollutants, a low concentration of SO_2 was consistently
255 measured throughout the whole study.

256 As shown in Figure 3, the concentrations of BC, NO_x , CO and SO_2 were highly
257 variable on the different routes in the NCP. The concentration ranges of these four
258 species were $5\text{--}14 \mu\text{g m}^{-3}$ for BC, $447\text{--}891$ ppb for NO_x , $22.6\text{--}40.4$ ppb for SO_2 and
259 $1105\text{--}1652$ ppb for CO. These extremely high values, i.e., 95 percentile
260 concentrations, were consistently found in various plumes near these emissions
261 hotspots in the NCP, which suggested a major influence on concentrations of
262 measured species in these hotspot areas by local emissions. The hotspots observed
263 here were mainly around the junction areas of our design routes, and they included but
264 were not limited to areas of Beijing, Tianjin, Baoding, Cangzhou, Dezhou,
265 Shijiazhuang and Zhuozhou. Previous model simulations and satellite measurements
266 in the NCP also confirmed the high concentrations of NO_2 around these large cities
267 (Shi et al., 2008). It is worth noting that these observed concentration hotspots moved
268 around the emissions hotspots, probably as a result of the varied transport processes in
269 different trips. For example, a pollution plume was detected 100 km to the south of

270 Cangzhou on June 20, but 130 km to the north of Cangzhou on July 6. In addition,
271 plumes were not always detected in different experiments around these cities, with the
272 exception of Shijiazhuang.

273 Figure 3 here.

274 During the five experiments, no clear temporal distributions of air pollutant
275 concentrations in the NCP were seen, except for the significantly low levels of NO_x
276 and SO₂ observed in the last experiments. However, no connections between the
277 decline in NO_x and SO₂ concentrations and emissions or transport could be made. In
278 fact, precipitation caused the low concentrations of NO_x and SO₂ in the last
279 experiment in NCP. And all measuring areas were affected by heavy rain, so the
280 concentrations of NO_x and SO₂ on all routes were low.

281 In summary, our mobile laboratory measurements indicated spatial distributions of
282 the pollutants SO₂, CO and BC and the number density of fine particles. The
283 concentrations of air pollutants in the NCP were among the highest in the world and
284 extremely high concentrations were also observed around several cities.

285 **3.2 The influence of traffic emission**

286 The levels and distributions of air pollutants in the NCP are mainly attributable to
287 three sources, i.e., regional transport, local emissions and traffic emissions. On-road
288 measurements, however, could be greatly affected by traffic emissions (Wang et al.,
289 2009). The influence of traffic emissions on our mobile laboratory measurements is

290 discussed below.

291 **3.2.1 1. Comparing the on- and off-road measurements of air pollutants to**
292 **estimate the enhancement of air pollution on highway above the regional**
293 **background**

294 Each on-road measurement trip started from a parking lot in a highway service center
295 and ended at the parking lot of another service center. The parking lots are about 150
296 m away from highway, such as those in service centers Dezhou (DZ) and Xizhaotong
297 (XZT) (Fig. S3). Using the difference of the concentrations of air pollutants measured
298 in a parking lot and on highway, we can estimate the level of the enhancement of the
299 concentrations of air pollutants on highway above regional backgrounds.

300 Table 2 shows the 5 min averaged concentrations of NO_x, CO, SO₂, and BC
301 measured in parking lots and on highways, and the difference between the on- and
302 off-road concentrations. The concentrations in parking lots were measured for 5 mins
303 before driving or after parking with engine turned off, and the concentrations on
304 highways were measured for 5 mins after entering highways or 5 mins before entering
305 the service centers.

306 The concentrations of NO_x measured on highway show drastic enhancement than
307 those measured off-road, from 19 to 449 ppb, or 43% to 1658% (510±61%, mean±
308 Standard deviation), while other pollutants have much lower enhancement or even
309 reduction in concentrations. The difference between the on-road and off-road
310 concentrations of CO ranged from -478 to 145 ppb, or -28% to 34% (7±22%); for SO₂,

311 it is -26 to 13 ppb or -18% to 175% ($52\pm 59\%$); for BC, it is -0.7 to 4.7 $\mu\text{g m}^{-3}$ or -11%
312 to 261% ($85\pm 90\%$).

313 The 175% enhancement of SO_2 on July 12 could be due to the much lower SO_2
314 concentration, 1.2 to 3.3 ppb, than those measured in other days. The rain on July 12
315 is likely the major reason for the much lower concentrations of SO_2 (3 ppb) and NO_x
316 (63 ppb), while CO and BC, which are much less water soluble, show no significant
317 difference than those measured in other days. If we ignore the results of SO_2 and NO_x
318 on July 12, then the difference between the on- and off-road concentrations were 82%
319 to 1658% ($510\pm 61\%$) for NO_x and were -18% to 87% ($31\pm 31\%$) for SO_2 .

320 Apparently, vehicular emission is the major source lead to the 82% to 1658%
321 enhancement of NO_x concentrations on highway. The enhancement of CO and SO_2
322 concentrations on highway were mostly less than 30%, suggesting vehicular emission
323 is not the main source for CO and SO_2 on highway, regional background is the
324 dominant factor determine their concentrations.

325 **3.2.2 2. Time series of the concentrations of air pollutants measured on highway**

326 Figure 4 shows the concentrations of NO_x , CO, SO_2 , and BC measured on highway
327 on a typical day, June 13, 2013. Apparently, the concentration of BC follows the trend
328 of NO_x concentration and those of CO and SO_2 did not. This suggests the high
329 enhancement of NO_x and BC concentration on highway was due to the vehicular
330 emission, while the concentrations of CO, SO_2 were not.

331 Figure 4. here.

332 **3.2.3 Using the ratios of weighted vehicular emission factors to estimate the**
333 **concentration enhancement of NO_x, CO, SO₂ and BC measured on highway**

334 Based on the reported vehicular emission factors (Shen et al., 2015; Cai and Xie, 2007,
335 2010; Lei et al., 2011) and the vehicle composition (Chinese Automotive Technology
336 & Research Centre, 2015) in Hebei province, where we conducted the most of the
337 mobile measurements, we estimated the weighted vehicular emission factors on the
338 highways (Table 3). The factors for NO_x, CO, SO₂, and BC during our measurements
339 were 2.9 g km⁻¹, 4.8 g km⁻¹, 0.04 g km⁻¹, and 0.01 g km⁻¹, respectively. If we assumed
340 400 ppb as the NO_x concentration enhancement on-road caused by vehicular emission,
341 using the ratios of the weighted vehicular emission factors, the estimated
342 enhancements of CO, SO₂ and BC concentrations on-road emitted by vehicles were
343 240 ppb, 3 ppb and 1 µg m⁻³. These are at the similar levels of those enhancement
344 showed in Table 1, suggesting vehicular emission is the main source for the
345 enhancement of the on-road concentrations of NO_x, CO, SO₂, and BC. However, the
346 enhancement of CO and SO₂ concentrations on highway were mostly less than 30%,
347 this provides further evidence that CO and SO₂ concentrations on highway were
348 dominated by regional background; and vehicular emission was not the main source.

349 Table 3. here.

350 **3.2.4 Correlations between NO_x, CO, SO₂ and BC with NO**

351 A strong correlation ($r^2 = 0.99$) was found between on-road NO_x and NO (Fig. 5),
352 with an average NO/NO₂ ratio of 4, which was much higher than the value of 0.05–

353 0.2 in the aged plumes (Finlayson-Pitts and Pitts, 2010). The results indicated that
354 NO_x observed by our mobile laboratory was mostly influenced by fresh vehicle
355 emissions. Overall, the on-road NO_x observations here were not representative of the
356 NO_x levels in the NCP. While, the concentrations of BC, CO and SO₂ were not
357 correlated with those of NO.

358 Figure 5. here.

359 3.2.5 Possible traffic jams and high concentrations of air pollutants

360 Each on-road measurement trip started from a parking lot in a highway service center
361 and ended at the parking lot of another service center. The driven speed of the mobile
362 platform was never lower than 80 km h⁻¹ on highways, this is one of the evidences
363 that we encountered no traffic jams, and the number of vehicles around our mobile
364 platform seldom increased.

365 During our measurements, as shown in Fig. 3, we always measured high
366 concentrations of air pollutants near large cities. As the time series of the
367 concentrations of NO_x, CO, SO₂ and BC measured on June 13 (Fig. 6) show, when
368 approaching Baoding, CO concentration gradually increased from 500 ppb at 50 km
369 away from Baoding to 1200 ppb at 23 km away from Baoding. The same is SO₂ (from
370 10 ppb to 40 ppb) and BC (from 4 μg m⁻³ to 8 μg m⁻³), while NO_x does not show the
371 same trend. When at 15 km away from Baoding, we see another increase of all the
372 pollutants, and the same decrease trend after at 10 km from Baoding. The same
373 continuously increasing trends of SO₂ and CO, started at 50 km to 15 km away from

374 Baoding, indicated the regional transport of SO₂ and CO. Started at 15 km, away from
375 Baoding, all the four pollutants had a peak with about 10 km width, which indicated
376 the urban emissions.

377 **Figure 6. here.**

378 In summary, both the comparison of on- and off-road measured concentrations and
379 the vehicular emission factors provided the evidences:

380 (1) Vehicular emission is the main source for the enhancement of the on-road
381 concentrations of NO_x, CO, SO₂, and BC;

382 (2) The high enhancement of NO_x concentration on highway suggesting NO_x on
383 highway was mainly from vehicular emission;

384 (3) CO and SO₂ concentration have up to 20% and 31% average enhancement on
385 highway, suggesting that CO and SO₂ on highway were mainly from regional
386 background.

387 (4) The difference in the enhancement is the difference of level of background
388 contribution. NO_x has shorter life time, while the lifetimes of CO and SO₂ in the
389 atmosphere are longer, and regional background contribution become important

390 Thus, the CO and SO₂ measurement results could be used for studying the spatial
391 distributions of the air pollutants in the NCP region. The hot spots measured near
392 large cities were mainly from local emissions.

393 **3.3 The influence of regional transport**

394 Local emissions and regional transport are the two main sources of pollutants in the

395 NCP (Xu et al., 2011). As stated above, local emissions in large cities had a major
396 impact on the air quality in their adjacent areas. We found all air pollutants measured
397 near large cities were at high level. According to the analyzation in section 3.2, they
398 were mainly from local emissions of cities. Also, according to the estimated emission
399 inventories, these cities were major sources of air pollution which caused the
400 concentrations of air pollution high around them. Regional transport also plays a
401 major role. Our study demonstrated that the contribution of regional transport could
402 vary both spatially and temporally, depending on a number of parameters, such as
403 prevalent wind, terrain and vertical mixing. We also roughly divided the NCP into two
404 parts according to these parameters, i.e., the northern border area and the central area,
405 to discuss the influence of regional transport on air quality. All trip-average
406 concentrations of SO₂, CO and BC and wind directions on those days discussed in the
407 manuscript were shown in table 4.

408 Table 4. here.

409 **3.3.1 The border areas of NCP**

410 The northern border area of the NCP included major parts of routes 4 and 5 and the
411 western border areas of the NCP included a major part of route 1. The area is
412 surrounded by the Taihang Mountains to the west and the Yan Mountains to the north.
413 The north wind prevailed in the winter and the south wind prevailed in the summer in
414 this area.

415 During the measurements, the three routes experienced both north and south winds.
416 Specifically, northwest winds and east winds brought outside air masses from
417 Northeast China and the Bohai Sea to the northern border area on July 2 (route 1) and
418 July 7 (route 5), respectively (Fig. 7). In both trips, the concentrations of SO₂, CO and
419 BC were 4.5 ± 2.3 ppb, 550 ± 240 ppb and $5.0 \pm 2.6 \mu\text{g m}^{-3}$, respectively, on July 2
420 and 7.0 ± 3.0 ppb, 1090 ± 320 ppb and $6.5 \pm 2.7 \mu\text{g m}^{-3}$, respectively, on July 7 (Fig.
421 2), which were the lowest values observed here in the border areas of the NCP. These
422 observations were reasonable as areas including the Bohai Sea to the west, north and
423 east of the NCP were regions of low emissions (Fig. 3) and the clean air brought by
424 northeast and east winds could dilute the air pollutants in the border areas of the NCP.

425 Figure 7 here.

426 It is worth noting that the BC concentration was not lowest on July 2, which was
427 the opposite of the observations for the gas pollutants, SO₂ and CO. Satellite images
428 showed that there were many fire plots near route 1 on July 2 (Fig. S4). A featured
429 single peak of aerosol number density at around 50 nm (Fig. S5) further confirmed
430 that BC emissions from agricultural crop residue burning contributed significantly to
431 the BC levels on July 2 (Zhang et al., 2011; Hays et al., 2005; Li et al., 2007).

432 On June 24 (route 1), June 14 (route 4) and June 15 (route 5), the air masses were
433 transported inside the NCP from the southern NCP to the northern border areas by
434 south winds (Fig. 7). Under these wind conditions, the concentrations of SO₂, CO and
435 BC were 15 ± 5.8 ppb, 1300 ± 330 ppb and $8.0 \pm 1.4 \mu\text{g m}^{-3}$, respectively, on June 24,

436 26 ± 7.9 ppb, 1200 ± 230 ppb and $6.5 \pm 1.5 \mu\text{g m}^{-3}$, respectively, on June 14 and $28 \pm$
437 7.1 ppb, 1600 ± 370 ppb and $7.0 \pm 1.9 \mu\text{g m}^{-3}$, respectively, on June 15 (Fig. 2), which
438 were among the highest levels detected on these routes. According to the emissions
439 inventories (Fig. 3), most emissions hotspots were located in the central and southern
440 parts of the NCP. Pollutants could be easily accumulated in air masses of south and
441 central NCP origin. In addition, the Yan Mountains to the north of the border area
442 stopped the possible transport pathway of these air masses, which further enhanced
443 the accumulation of long-lived air pollutants in the northern border area of the NCP.

444 Although route 4 on July 6 was under the influence of south winds, as the same
445 route on June 14 (Fig. 7), the concentrations of SO_2 and BC on July 6 were 14 ± 7.6
446 ppb and $4.6 \pm 1.8 \mu\text{g m}^{-3}$, respectively, on July 6, which were much lower than the
447 values of 26 ± 7.9 ppb and $6.5 \pm 1.5 \mu\text{g m}^{-3}$, respectively, on June 14. Meanwhile, the
448 CO levels on these two days were similar. One possible cause of the low
449 concentrations of both SO_2 and BC was the slightly higher boundary layer height on
450 July 6 compared to June 14. The precipitation that occurred on July 6, but not on June
451 14, appeared to be a more important contributing factor. The solubility of CO is less
452 than that of SO_2 . Therefore, the wet deposition lifetime of SO_2 would be much shorter,
453 thus limiting the transport distance of SO_2 . Meanwhile, the wet deposition of BC
454 particles would also prohibit its long-range transport. This may explain the similar CO
455 levels and low SO_2 and BC levels on July 6 at the same time.

456 In conclusion, for the northern border area, local emissions and regional transport

457 from other NCP areas due to south winds were two main sources of long-lived
458 pollutants; both north and east winds had significant dilution effects on the
459 concentrations of gas pollutants. It was based on a general situation that the air quality
460 in the north border of the North China Plain was clean, because the emissions of air
461 pollutants were low in that area (Fig. 3). The wind dependency scatter plots for SO₂
462 were used to show the contribution of regional transport to air pollution in the
463 northern border area of the NCP (Fig. 8). The results indicated that the high
464 concentration was connected to the south wind at a wind speed from 4 to 10 m s⁻¹.
465 Similar results for SO₂ and CO were reported for several sites in the northern NCP
466 (Wu et al., 2011; Lin et al., 2011; Lin et al., 2009). As the south wind usually
467 prevailed in the summer and north wind in the winter, the regional transport of
468 long-lived pollutants within the NCP from the central and southern parts to the
469 northern parts should be prevalent in the summer; while the dilution of air pollutants
470 mainly by north winds and occasionally east winds should be prevalent for the
471 northern parts of the NCP in the winter.

472 Figure 8 here.

473 3.2.2 The central NCP

474 The central NCP consisted of routes 2 and 3, where numerous heavily polluted cities
475 are located. The area was surrounded by the Taihang Mountains to the west or
476 emissions hotspots in other directions. While the north wind prevailed in the winter, as
477 for the northern border areas, low pressure prevailed in the summer with south and

478 northeast winds in this area.

479 During the observation period, the measurements along the two routes experienced
480 different wind fields. Route 2 experienced southwest winds on June 12 and July 3,
481 northeast winds on June 18 and a low-pressure system with south and northeast winds
482 on June 25 (Fig. S6). Unlike the northern border area of the NCP where strong north
483 winds had a dilution effect, the concentrations of gas pollutants were mostly high
484 regardless of the wind direction on route 2 in the central NCP, e.g., on June 18 and
485 July 3 (Fig. S6). Generally, our observations were reasonable according to the unique
486 terrain and emissions map in central NCP. Due to the heavy emissions level in the
487 central NCP and surrounding areas, pollutants readily accumulated to high levels on
488 their ways to the central NCP in air masses from all directions, such as the clean air
489 masses from the Bohai Sea, West China and Northeast China, and polluted air masses
490 from Southeast China.

491 The situation was slightly different in areas along route 3, particularly for those off
492 the coast of the Bohai Sea. Route 3 experienced east winds on July 14 (Fig. S6), and
493 the concentrations of pollutants were low (Fig. 2). This was not only because of the
494 wet deposition from the rain on that day, but also the transport of clean air from the
495 Bohai Sea. A featured peak of aerosol number density at around 20 nm (Fig. S5)
496 further confirmed the incoming air from the Bohai Sea (Haaf and Jaenicke, 1980;
497 Hoppel et al., 1986).

498 Vertical mixing can also affect the concentrations of pollutants. For example, while

499 the wind fields were similar on route 3 on June 13 and July 4 (Fig. 9), the
500 concentration of pollutants on June 13 was lower than that on July 4 (Fig. 2). This was
501 because the boundary layer was much higher on June 13 (976 m) than on July 4 (626
502 m), and the strong vertical convection diluted the air pollutants.

503 Specifically, the relative contributions of emissions and regional transport to the
504 local air pollution levels were slightly different on different routes. At the junction of
505 routes 1 and 2 around Shijiazhuang area, the concentrations of air pollutants were
506 always high, except in the last experiment when wet precipitation occurred. The local
507 emissions contributed significantly to the air pollution levels in this area. The city of
508 Shijiazhuang is known as an emissions hotspot with heavy coal consumption.
509 Previous model result showed that Shijiazhuang is an important emissions hotspot of
510 SO₂ even in the whole NCP area (He et al., 2012). Meanwhile, the Taihang Mountains
511 to the west of the city prevented the diffusion of air pollutants. On the other hand,
512 transport convergence in front of the Taihang Mountains and Yan Mountains was
513 proposed in a previous study (Su et al., 2004), as a result of a low-pressure system
514 along the Taihang Mountains and Yan Mountains. Although the transport convergence
515 moved around in the north end, it always passed by the Shijiazhuang area. As shown
516 in Figure 3, high levels of pollutants, particularly relatively long-lived species such as
517 SO₂ and CO, were consistently observed at the western end of route 2 near
518 Shijiazhuang area. Broad peaks of SO₂ and CO concentrations, indicators of regional
519 transport plumes, were present near Shijiazhuang area (Fig. S7).

520 Similar to the situation in Shijiazhuang area, transport convergence would
521 occasionally pass through other cities along routes 2 and 3. In a typical case on June
522 13 (Fig. 9), air masses were transported far from the southwest of the NCP along the
523 transport convergence through Shijiazhuang and reached the area on route 3, with air
524 pollutants accumulating during transport and showing high concentrations.

525 Figure 9 here.

526 Overall, in most areas in central NCP, regional transport would play essential roles
527 in determining the local air pollution levels, although the underlying mechanisms
528 were different for the transport convergence area, central NCP area and coastal area.
529 The wind dependence scatter plots for SO₂ and CO were used as examples to show
530 the contribution of regional transport on air pollution in central NCP (Fig. 8). The
531 results indicated that the prevailing winds were southwest and northeast in central
532 NCP during our observation period. The concentrations of CO were independent of
533 wind direction and wind speed. In addition, the high concentrations of SO₂ were
534 related to southeast winds with high speed, which was about 5–10 m s⁻¹. This may
535 have been because of transport convergence. Due to the strong interaction of different
536 areas in central NCP, emissions control policies must consider the whole emissions
537 budget to achieve the air quality aims.

538 We discussed five impactors: local emission, precipitation, location, wind direction
539 and boundary layer height. The influence of local emission reflected in the spatial
540 distribution of concentrations (Fig. 3). Hot spots were found near cities. However, for

541 route-average results (Fig. 2), local emission plays a minor role in the distribution of
542 concentrations, because the routes were mainly in suburb. The large reduction in SO₂
543 and NO_x concentration measured on July 12 along all routes were caused by
544 precipitation. The persistent high concentrations of pollutants in route 2 were
545 associated with the location of it, which was surrounded by high emission areas.
546 Besides the route 2, the different concentrations in one route in different
547 measurements were mainly from different wind directions. And we also found one
548 case that the concentrations changed lot between June 13 and July 4 in route 3 under
549 the similar wind directions. Based on the model results, boundary layer height might
550 explain the change.

551 **4. Conclusion**

552 A mobile laboratory was employed to obtain snapshots of the spatial distributions of
553 air pollutants in the NCP. The concentrations observed were at the highest levels in
554 the world and were distributed unevenly in the NCP. Most high concentrations, i.e., 95
555 percentile concentrations, of air pollutants were found near emissions hotspots, which
556 suggested the influence of local emissions. However, regional transport of air
557 pollutants was also considered significant in determining the air quality in the NCP.
558 Back trajectory analysis, satellite data and tracer pollutants were combined to
559 recognise various cases of regional transport in both the northern border and central
560 NCP. Where the border areas would occasionally be diluted by winds from outside the
561 NCP, the central NCP was affected by regional transport of air pollutants with a few

562 exceptions, such as when precipitation occurred. To achieve the aims of air quality
563 locally, emissions control policies must consider the whole emissions budget in the
564 NCP.

565

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567 The English in this document has been checked by at least two professional editors,
568 both native speakers of English. For a certificate, please see:

569 <http://www.textcheck.com/certificate/bENNRx>

570 **Data availability.** The data of mobile and stationary measurements are available upon
571 requests. Also, we are working on installing a web site in this year. We will provide
572 data of GPS, vehicle speed, meteorology, concentrations of air pollutants.

573 **Author contribution.** T. Zhu, Y. Zhu, Y. Han and W. Chen designed the experiments.
574 T. Zhu secured the research grants. Y. Zhu, Y. Han and W. Chen carried out the
575 experiments. J. Zhang developed the model code and performed the simulations. J.
576 Wang managed the data in the program. J. Liu provided the emission maps. L. Zeng,
577 Y. Wu, X. Wang, W. Wang and J. Chen provided the data of stationary measurements.
578 Y. Zhu analyzed the data with contributions from all co-authors. Y. Zhu prepared the
579 manuscript with helps from T. Zhu, C. Ye and Y. Li.

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789

790 Table 1. Precisions and uncertainties of air pollutants analyzers used in our
 791 experiments

	NO _x	CO	SO ₂	BC
Precision	1%	1%	0.5%	0.1 μg m ⁻³
Uncertainty	<10%	<100 ppb	<10%	<1%

Time resolution 30 s 40 s 120 s 6 s

792

793 Table 2. The concentrations of NO_x, CO, SO₂ and BC measured in parking lots and
 794 high ways, and the concentration differences between off-road and on-road
 795 measurement.

Date	Parking lot	Concentrations							
		NO _x (ppb)				CO (ppb)			
		Off-road (C1)	On-road (C2)	C2-C1	$\frac{C2-C1}{C1}$	Off-road (C1)	On-road (C2)	C2-C1	$\frac{C2-C1}{C1}$
6/11	XZT	234	643	409	175%	694	677	-17	-2%
6/12	XZT	411	699	288	70%	1050	1098	48	5%
6/12	DZ	20	106	86	430%	396	506	110	28%
6/13	DZ	19	334	315	1658%	431	576	145	34%
6/17	XZT	30	297	267	890%	1000	1086	86	9%
6/18	XZT	109	468	359	329%	1212	955	-257	-21%
6/18	DZ	372	677	305	82%	588	711	123	21%
7/3	XZT	49	498	449	916%	826	861	35	4%
7/12	XZT	44	63	19	43%	1708	1230	-478	-28%

796 Table 2 (continuous). The concentrations of NO_x, CO, SO₂ and BC measured in
 797 parking lots and high ways, and the concentration differences between off-road and
 798 on-road measurement.

Date	Parking lot	Concentrations							
		SO ₂ (ppb)				BC (μg m ⁻³)			
		Off-road (C1)	On-road (C2)	C2-C1	$\frac{C2-C1}{C1}$	Off-road (C1)	On-road (C2)	C2-C1	$\frac{C2-C1}{C1}$
6/11	XZT	11.3	12.7	1.4	12%	2.3	5.6	3.3	143%
6/12	XZT	24.4	30.9	6.5	27%	2.9	6.6	3.7	128%
6/12	DZ	8.9	10.0	1.1	12%	1.5	4.1	2.6	173%
6/13	DZ	14.6	12	-2.6	-18%	2.3	3.6	1.3	57%
6/17	XZT	11	17	6	55%	3.4	5.5	2.1	62%
6/18	XZT	15	28	13	87%	6.6	5.9	-0.7	-11%
6/18	DZ	17	21	4	24%	7.4	10.3	2.9	39%
7/3	XZT	17	21.9	4.9	29%	1.8	6.5	4.7	261%
7/12	XZT	1.2	3.3	2.1	175%	5.3	6.2	0.9	17%

799 Table 3. Estimated weighted vehicular emission factors of CO, NO_x, SO₂ and BC
 800 during mobile measurements in Hebei Province, China.

Vehicle type	Composition	Emission factors			
		CO ^[1] g km ⁻¹	NO _x ^[1] g km ⁻¹	SO ₂ ^[2] g km ⁻¹	BC ^[3] g km ⁻¹

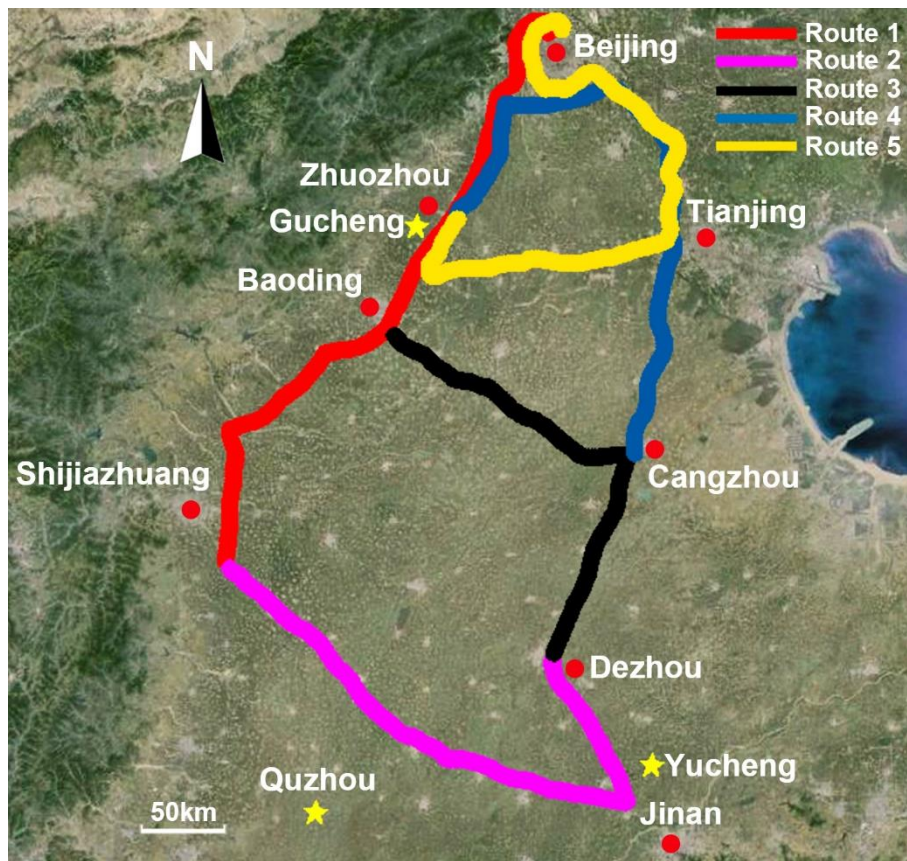
Diesel bus	20%	1.2	11	-	-
Medium-duty diesel vehicles	20%	1.5	6.4	-	-
Light-duty gasoline vehicles	50%	4.4	1.3	-	-
Heavy-duty vehicles	10%	1.6	6.6	0.4	0.11
Weighted emission factor		2.9	4.8	0.04	0.01

801 ^[1] Shen et al., 2015; ^[2] Cai and Xie, 2007; ^[3] Lei et al., 2011

802 Table 4. Trip-average concentrations of SO₂, CO and BC and wind directions on those
803 days discussed in the manuscript

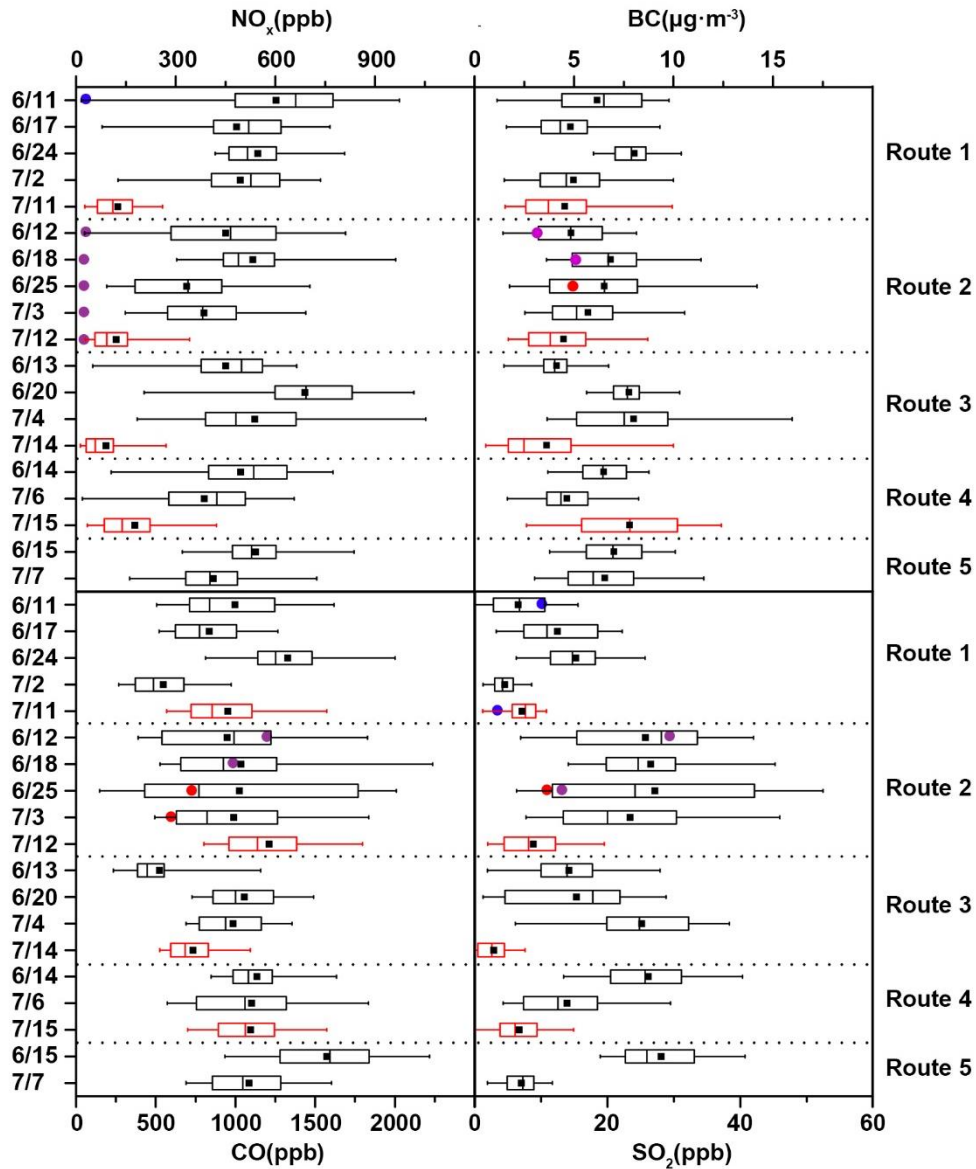
Route	Date	Concentrations (mean ± SD, ppb)			Wind directions
		SO ₂	CO	BC	
Route 1	July 2	4.5±2.3	550±240	5.0±2.6	northwest
Route 5	July 7	7.0±3.0	1090±320	6.5±2.7	east
Route 1	June 24	15±5.8	1300±330	8.0±1.4	south
Route 4	June 14	26±7.9	1200±230	6.5±1.5	south
Route 5	June 15	28±7.1	1600±370	7.0±1.9	south
Route 4	July 6	14±7.6	1100±450	4.6±1.8	south
Route 2	June 12	26±11	950±440	4.8±2.2	southwest
Route 2	June 18	27±10	1030±530	6.8±2.3	northwest
Route 2	June 25	27±16	1020±680	6.5±3.3	stable
Route 2	July 3	23±15	989±450	5.7±2.6	southwest

804



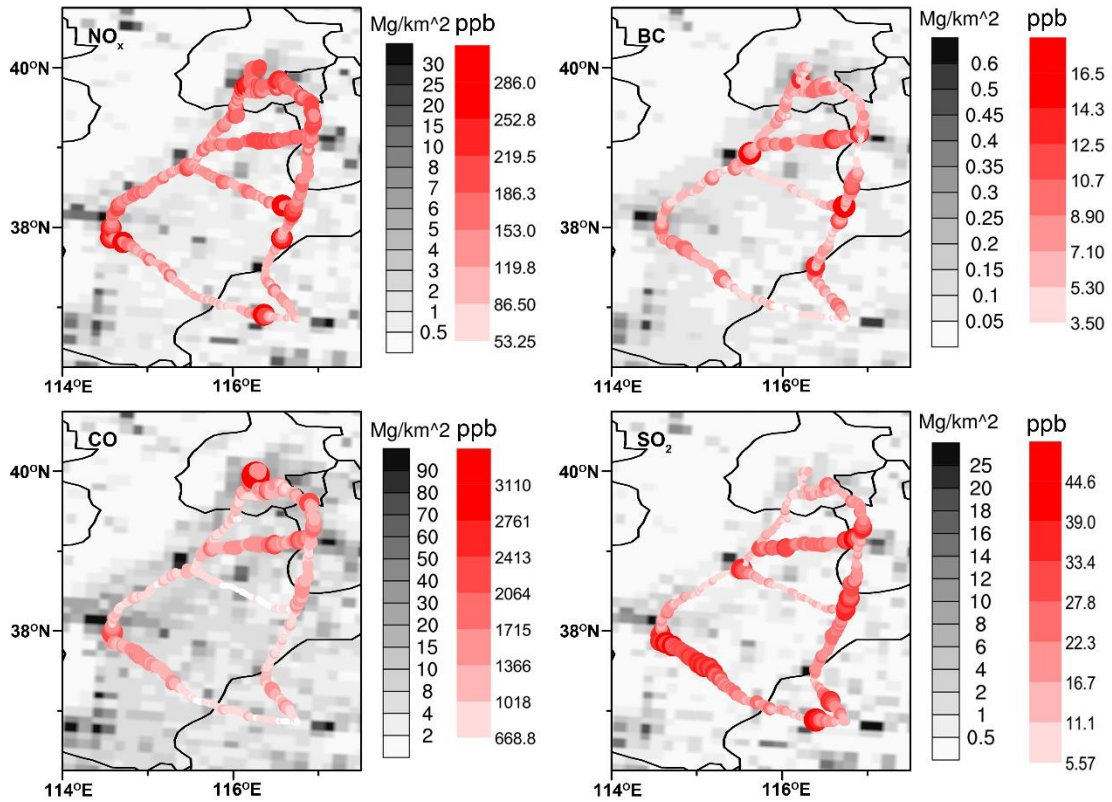
805

806 **Fig. 1.** The study area in NCP. The red track shows route 1, from Beijing to Shijiazhuang. The
 807 purple track shows route 2, from Shijiazhuang to Dezhou. The black track shows route 3, from
 808 Dezhou to Baoding. The blue track shows route 4, from Cangzhou to Zhuozhou. The yellow track
 809 shows route 5, from Zhuozhou to Beijing. The red round dots on the map present the major cities
 810 near the routes. The yellow five-pointed stars present the monitoring sites.



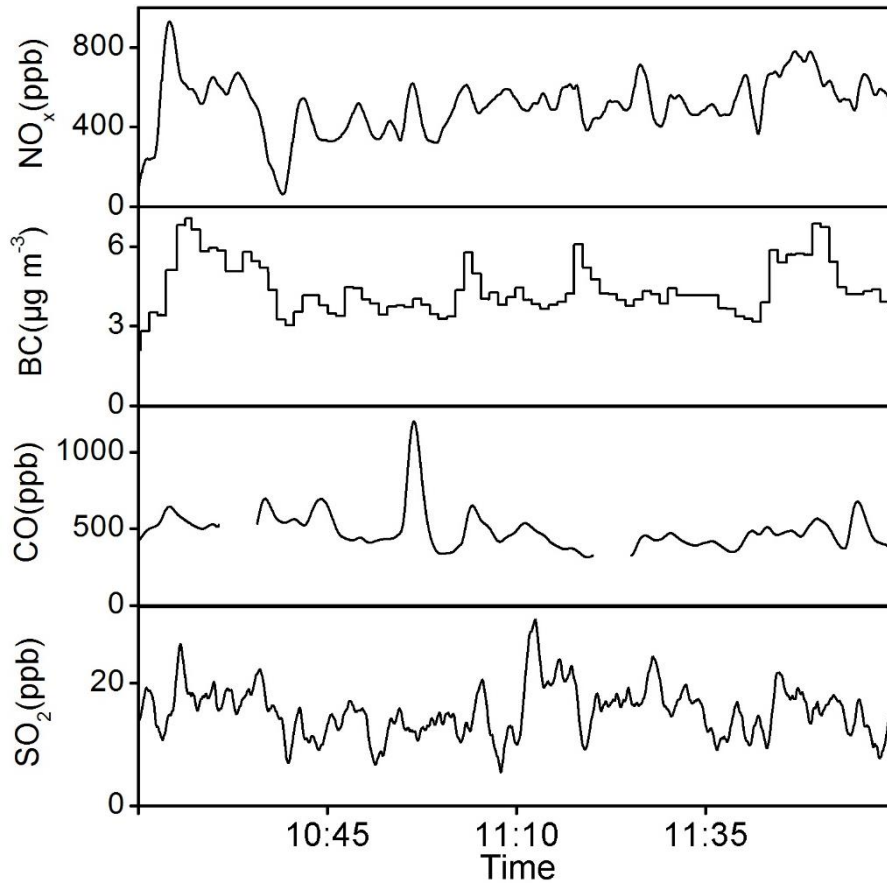
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812 **Fig. 2.** The concentrations of SO_2 , CO , BC and NO_x in each trip in different routes. The red boxes
 813 were the results in the last experiment. Values marked were the 5th and 95th percentile (-), standard
 814 deviation (lower and upper box lines), median (middle box line), and mean (■). The blue dots
 815 were results of GC station. The purple dots were results of QZ station. And the red dots were
 816 results of YC station.



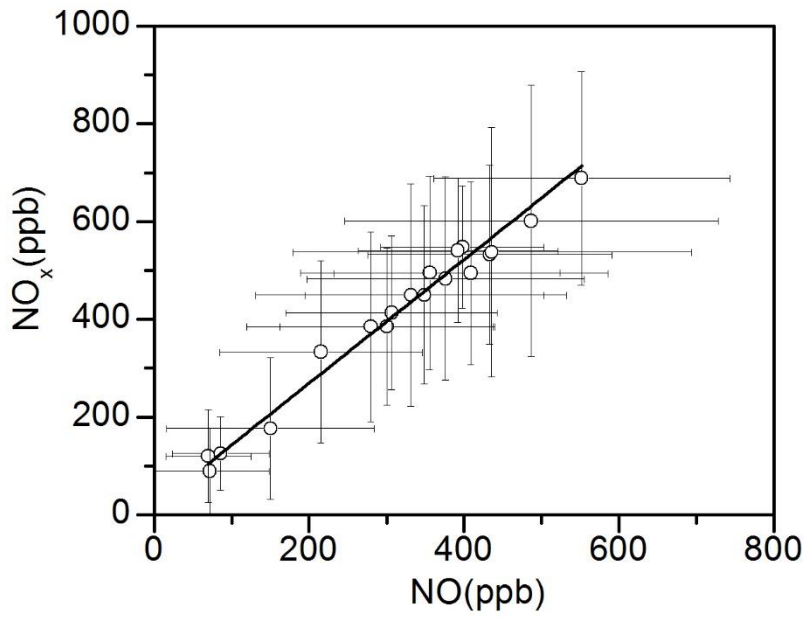
817

818 **Fig. 3.** The spatial distributions of the measured concentrations in our study and the emission
 819 maps of SO_2 , NO_x , CO and BC. The colored tracks were average concentrations measured in this
 820 mobile observation. The black and white maps were emission maps of the year 2010 derived from
 821 MEIC model (Zhao et al, 2013).



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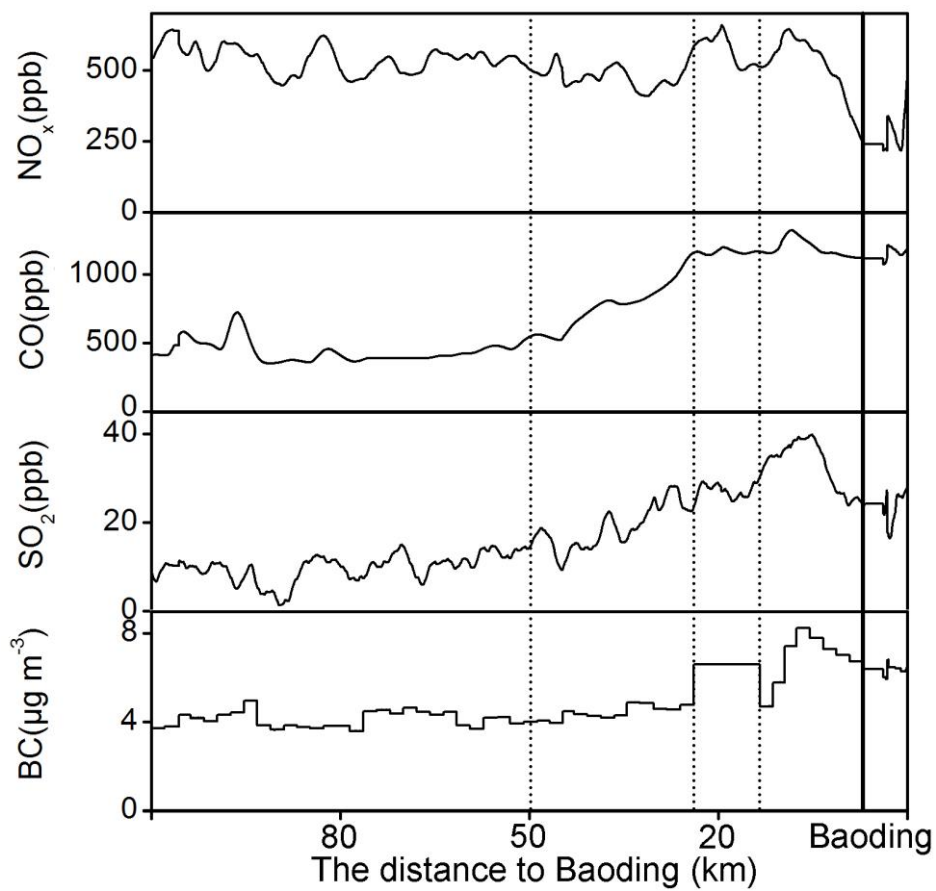
823 **Fig. 4.** The time series of the concentrations of NO_x , CO, SO_2 and BC measured on June 13.



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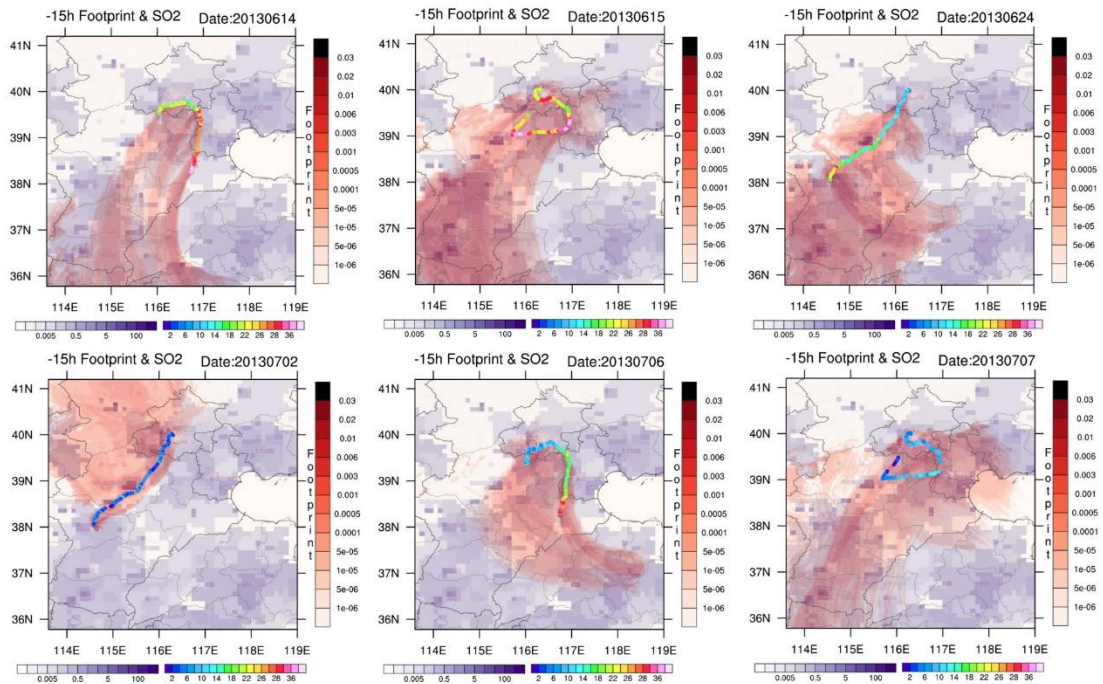
825 **Fig. 5.** The regression curve of the means of concentrations of NO_x and NO and the error bars in

826 all 19 trips.



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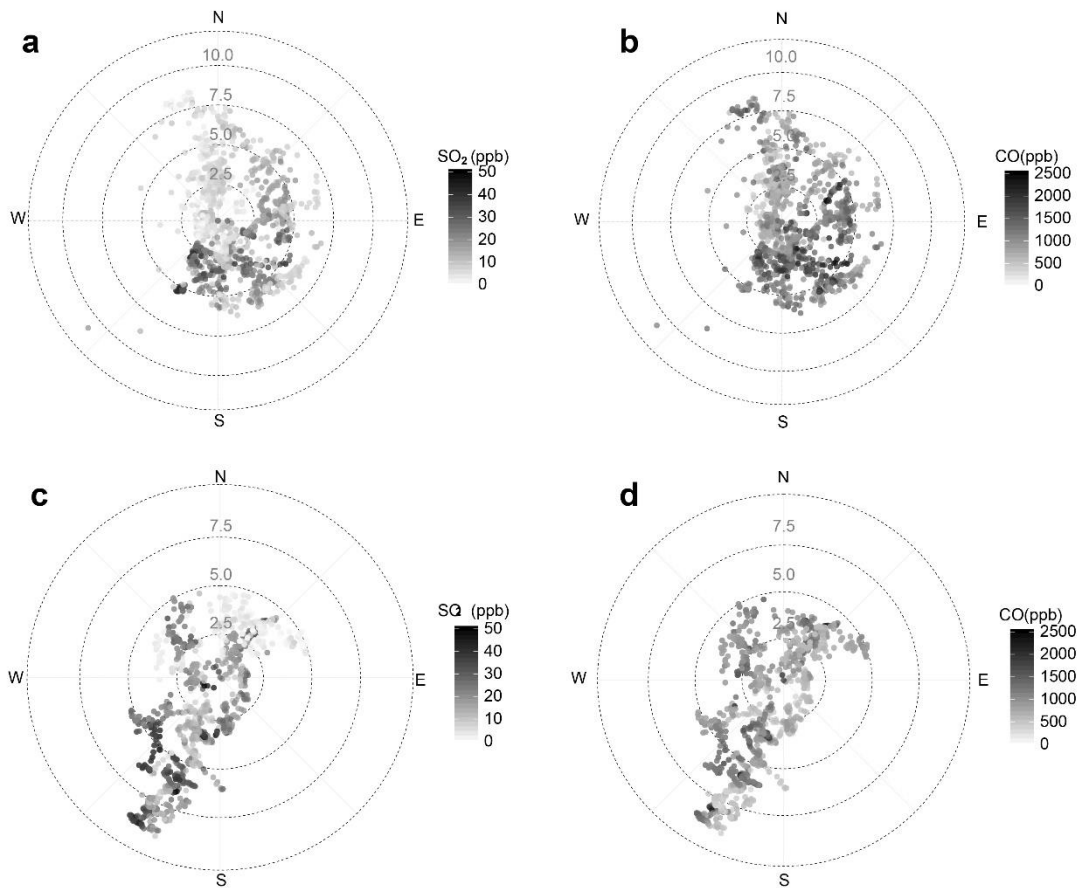
828 **Fig. 6.** The time series of the concentrations of NO_x , CO, SO_2 and BC measured on June 13 (the
829 four dashed lines marked the 50 km, 23 km, 15 km to Baoding and Baoding).



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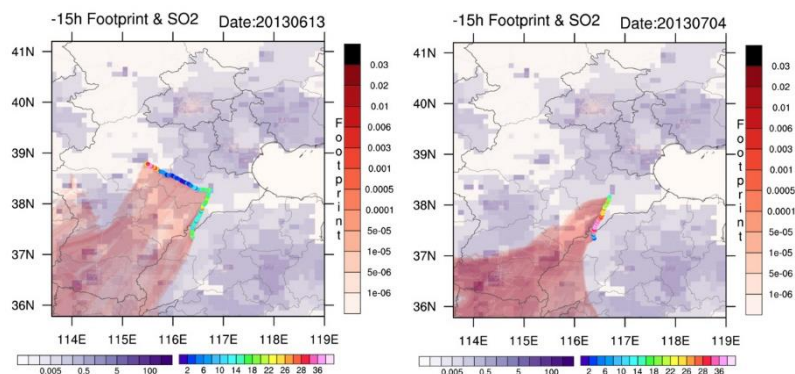
831 **Fig. 7.** The back trajectories of observed air masses in the borders of NCP in June 14, June 15,

832 June 24, July 2, July 6 and July 7.



833

834 **Fig. 8.** Wind dependency scatter plots of concentrations of SO₂ and CO in border and central areas
835 in NCP (a. SO₂ in border area; b. CO in border area; c. SO₂ in central area; d. CO in central area).



836

837 **Fig. 9.** The back trajectories of observed air masses in the central NCP in June 13 and July 4.

838