Response to the Comments of Referees

Observation of regional air pollutant transport between the megacity Beijing and the North China Plain

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We thank the referees 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: *The authors presented an analysis of the regional transport flux of pollutants between Beijing and North China Plain (NCP) based on two-years measurements at a single ground site (Yufa) located between the two regions. They also discussed a range of factors that contributed to the pollutant transport. The manuscript is concise and clear. However, the paper has major structure problems. While only four figures are included, the main text has extensive discussions on figures from the supplementary materials. Supplements are meant to provide information to readers, which is not key component to complete the paper. The authors need to reconsider what figures to include in the main text and avoid long discussions on supplements.*

Response: Accepted. This study provides a new approach based on long-term measurement at a cross-boundary site between Beijing and area southern to it to evaluate regional air pollution transport. To provide substantial information, we have restructured the manuscript, added 6 more figures in the main text and also expanded discussions to the relevant figures according to the referee's suggestion.

Changes in Manuscript: Figure 3-4, 6-7 and 9-10 have been added. Please refer to the revised manuscript, in Page 42-44, 46-47, and 49-50; we also added more discussion relevant to the

figures, please refer to the revised manuscript Section 3.2.2 from Page 17 Line 369 to Page 22 Line 474 for the discussions to Fig.6-7 and Section 3.3.2 Page 26 Line 567 to Page 27 Line 595 for the discussions to Fig.9-10.

Comment NO.2: *The paper lacks in-depth discussions and fails to present sufficient evidence to back their interpretations of the results (see details below). The transport flux calculation method comes with assumptions, and is subject to uncertainties and errors, which should be made clear to readers. The authors did not discuss at all how these assumptions and errors would affect the interpretation of results. I believe that the paper needs substantial revisions in order to be considered suitable for publication at ACP. Thus, I recommend rejecting the paper at this stage. The authors may consider a resubmission.*

Response: Agree to the revision suggestion. We have made substantial revision according to this comment. We added more description about the transport flux calculation method, the relevant assumption, the discussion about the uncertainty and errors, and PSCF analysis.

Changes in Manuscript: We have added more description about the transport flux calculation method and assumption, please refer to the revised manuscript in Section 2.2.2, from Page 9 Line 194 to Page 11 Line 231.

We have added more detailed information of the precisions and uncertainties of the measurement, please refer to the revised manuscript, from Page 7 Line 136 to Page 8 Line 158.

The uncertainty analysis was added in Section 3.3.3, please refer to the revised manuscript from Page 27 Line 596 to Page 28 Line 602.

The PSCF analysis based on HYSPLIT model was added to back our results of regional transport influence of Beijing and the NCP on the Yufa site. Please refer to the revised manuscript in section 2.2.3 from Page 11 Line 232 to Page 12 Line 262, section 3.3.2 from Page 26 Line 567 to Page 27 Line 595, and also Fig. 9 and 10 in Page 49-50.

Comment NO.3: *The authors use measurements at a certain height from a single round station*

to infer the transport flux between two vast regions – Beijing and NAP. The flux calculation method has many assumptions, which are not mentioned at all. Hourly winds are used in Eq. 3. Is it hourly mean and is the wind assumed to be constant? Note that winds can be highly variable within an hour. Are there any changes in wind speeds along the transport route between Beijing and NAP but not recorded at Yufa site? What are the conditions of the atmospheric boundary layer (stable, well mixed?) during the different years and seasons? The ABL condition strongly affects the pollutant mixing and vertical profile, and is expected to change a lot during different seasons (e.g., summer vs. winter). How are the seasonal PBL conditions accounted for in calculating the flux? Since the Yufa site is built on the top of a building, does the building or nearby structure affect the boundary layer and consequently, the wind and gas measurements? In addition, the extrapolation from Yufa site to the entire region is not backed by any analysis (e.g., trajectory analysis in HYSPLIT or STILT). How do the authors know that data collected at Yufa are representative for the entire region?

Response: Agree. The referee's concerns in this comment are responded below:

(1) Time resolution of the wind speed and air pollutant concentration

The flux intensity reported in the manuscript is the product of wind speed and air pollutant concentration measured at the same location. Ideally, we need to use the wind speed and air pollutant concentration with infinite small time resolution to conduct the surface flux calculations. In this study, the hourly data of the pollutants and wind were used, mainly because the pollutants concentration data was converted from the minutes' data to hourly mean to remove the accidental fluctuation and reduce the noise. Therefore, we assumed the wind speed and wind direction were constant within one hour, and hourly wind data were used to match with the hourly air pollutant concentration data to calculate the flux intensity.

(2) The spatial representation of the Yufa site

The Yufa site locates in the temperate monsoon climate zone and the topography of the surrounding area is flat (Fig.1 in the revised manuscript), the prevailing wind is with the same as

the surrounding region (Lin et al., 2009). Thus the wind field of the Yufa site is representative in the researched area in this study.

(3) The spatial representative of the flux intensity

The novelty of our study is to develop a method based on long-term ground based measurement to calculate the surface transport flux intensity across a cross-boundary site. We need to make it clear that the surface flux intensity calculated in this study is the per unit area flux across the Yufa site, which is different from the flux across a large area reported in other studies (e.g. Wang et al. 2011). Our results could only be extrapolated if the concentrations of all the pollutants and wind speed and direction were homogenously distributed, vertically and horizontally. Otherwise, we need vertical profiles of air pollutants concentration and wind to calculate the cross-section transport flux of two adjacent regions for the whole boundary layer with the integrating formula: $FLUX = \iint C_{(x,z)} WS_{(x,z)} sin\theta_{(x,z)} dxdz = \iint f_{(x,z)} dxdz$; where x is horizontal distance to the observed point, z is the vertical distance from ground to the observed point. In this study, we only focus on the method developing and evaluation of the regional transport influence of Beijing and the NCP on the cross-boundary site based on the ground-based observation data.

(4) The extrapolation from Yufa site to the entire region

In this study, we did not intent to extrapolate from Yufa site to the entire region. We only focus on the method developing and evaluation of the regional transport influence of Beijing and the NCP on the cross-boundary site based on the ground-based observation data. We conducted Bivariate Polar plots analysis and surface flux intensity calculation and get evidence of surface pollutants transport from Beijing to the Yufa site and from the NCP to the Yufa site. Considering the variations of the vertical and horizontal distributions of the air pollutants and meteorological parameters, and influence of the boundary layer on the regional transport, more multidimensional data with high precision and resolution are needs for further comprehensive discussion of the regional transport between Beijing and NCP.

(5) The Yufa site building height

As the Yufa site located on the top of a building (about 20 m above ground level) on the campus of Huangpu College. There is no tall building around the Yufa site which affects the wind and gas measurements. Meanwhile the site is only 20 m above ground level, so it is not likely affect the boundary layer (Cheng et al.,2001; He et al., 2006; Wang et al., 2012).

Changes in Manuscript: The discussion relevant to (1) Time resolution of the wind speed and air pollutant concentration and (3) spatial representative of the flux intensity were added, please refer to the revised manuscript in Section 2.2.2, from Page 9 Line 194 to Page 11 Line 231; the discussion relevant to (2) The spatial representation of the Yufa site and (5) The Yufa site building height were added in Section 2.1, please refer to the revised manuscript from Page 6 Line 125 to Line 131; the discussion relevant to (4) The extrapolation from Yufa site to the entire region, please refer to the revised manuscript from Page 27 Line 603 to Line 612.

Comment NO.4: *L53 and other places: avoid putting citations in the middle of a sentence. Put them at the end.*

Response: Accepted.

Changes in Manuscript: We have checked the manuscript and moved the citations to the end of the sentence in the revised manuscript. Please refer to the revised manuscript, in Page 4 Line 69 and Page 5 Line 90.

Comment NO.5: *L67: need to define the abbreviations (SO_2, CO……) (write their full names). Only need to define abbre. at their first occurrence. On L119, SO_2 is defined, but at the wrong place.*

Response: Accepted.

Changes in Manuscript： We have defined the abbreviations (SO_2, NO, NO_2, NO_x, CO, O_3 and O_x) at the right position in the revised manuscript. Please refer to the revised manuscript, from

Page 4 Line 80 to Line 82.

Comment NO.6: *Figure 1: The authors need to make the best use of figures. Fig. 1 is too simple and does not convene much information. I recommend placing marks or better, using color coding, to show where exactly NAP is located at. I suggest using terrain height as the background in Fig.1, since topography is a big factor in affecting the air quality in Beijing.*

Response: Accepted.

Changes in Manuscript: Figure 1 was revised following the referee's suggestion.

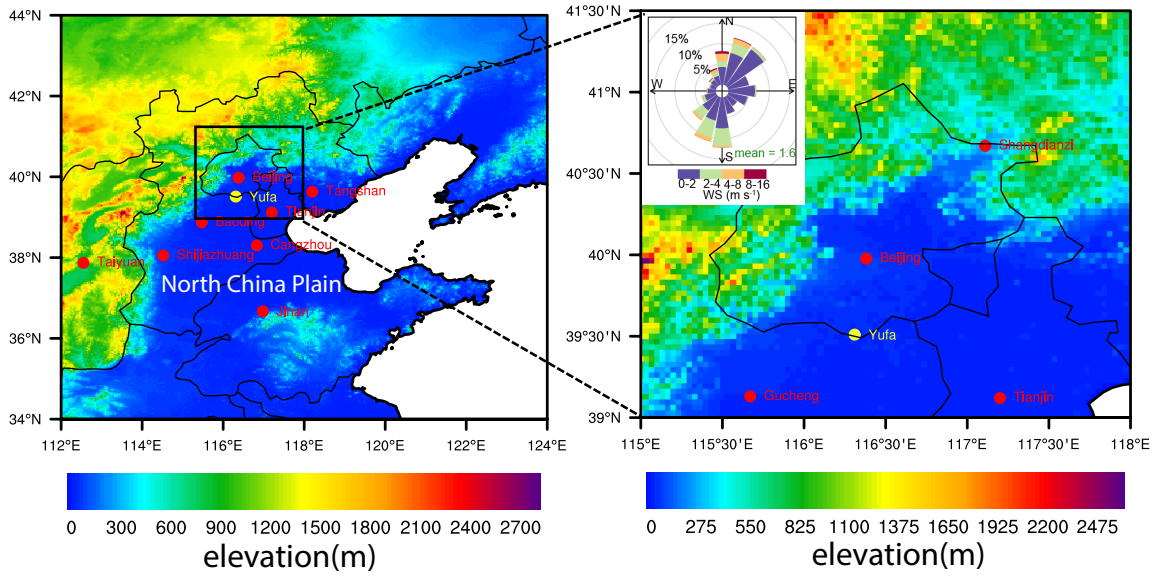

Figure 1. Overview information of the Yufa site.

Comment NO.7: *Sect. 1.1: what are the instrument accuracies for the gas species? Eq. 2: what does the smooth function do? L159: citation for White et al is missing. Double check that the reference is complete. L169: haven't you already described the angle at L165? Eq. 3: how is the cross section area calculated?*

Response: Accepted. We have added more detailed information of the instrument in the revised manuscript and listed it in Table 1.

Table 1. The information of the measurement instruments.

Species/ Parameter	Instrument	Detection limit	Time resolution	Precision	Uncertainty
SO_2	Ecotech 9850B	0.3 ppb	1 min	0.5% (1ppb)	5%
NO-NO_x	Ecotech 9841B	50 ppb	1 min	1% (0.5ppb)	10%
CO	Ecotech 9830	0.4 ppb	1 min	1% (0.1ppm)	1%
O_3	Ecotech 9810B	0.5 ppb	1 min	0.5% (1ppb)	5%
WS	LASTEM	-	10 min	0,1 m s^{-1}	5%
WD	LASTEM	-	10 min	0,1 º	1%
BP	LASTEM	-	10 min	0,1 hPa	±0.35 hPa
T	LASTEM	-	10 min	0,1 ℃	±0.2°C
RH	LASTEM	-	10 min	1%	±3%

The 24h smoothing is conducted by a smooth averaging method to remove the daily variations of the data which is equivalent to a low volume filtering. From the smoothing line we can easily recognize the “saw-teethed” variations of the pollutants between low and high concentrations and find the cycling of the clean and polluted stage. The cross section is the assumed unit area above the Yufa site at arbitrary height.

Changes in Manuscript: We have added more detailed information of the precisions and uncertainties of the measurement, please refer to the revised manuscript, from Page 7 Line 136 to Page 8 Line 158; the missing citation for White et al. was added, please refer to the revised manuscript, from Page 35 Line 784 to Line 786; The repeated description of the angle was deleted.

Comment NO.8: *L191: define WS, RH, TF and BP. Consider moving Fig. S2 to main text because you talk about it a lot! Sect. 3.2, L226-234, move the description on bivariate polar plots to the methods section.*

Response: Accepted.

Changes in Manuscript: The definition of WS, RH, T and BP were given in the right place, please refer to the revised manuscript from Page 7 Line 151-153. Fig. S2 and Fig. S3 was moved to the main text as Fig. 3 and Fig.4, please refer to the revised manuscript from Page 42 to Page 44;

the description of on the bivariate polar plots was moved to the methods section, please refer to the revised manuscript from Page 9 Line 187 to Line 193.

Comment NO.9: *L278-322: I suggest splitting the discussions on local emissions vs transport by different seasons for different species, because on one hand the emission source has a seasonal cycle and on the other hand, the seasonal meteorological conditions affect the chemical reaction rate, species lifetime and transport. I feel that this part of discussions is not backed by any solid analysis at all, but it is key to understand why different species behave differently on the bivariate polar plot.*

Response: Agree.

Changes in Manuscript: We have added Fig.7 and more corresponding discussion in section 3.2.2 on the seasonal variations of bivariate polar plots for different species. Please refer to the revised manuscript from Page 17 Line 369 to Page 22 Line 474.

Comment NO.10: *L307: SO_2 lifetime in the atmosphere is typically a couple of hours to 1-2 days.*

Response: Accepted.

Changes in Manuscript: We have corrected SO_2 lifetime in the atmosphere to a couple of hours to 1-2 days, please refer to the revised manuscript from Page 20 Line 431.

Comment NO.11: *Sect. 3.3: the fact that the flux values in Table 1 do not come with standard deviations points to the lack of uncertainty analysis in the flux calculations. Table 1 is subject to various errors which should be discussed and addressed.*

Response: Accepted.

Changes in Manuscript: The standard deviations were added for the mean surface flux intensities in Table 2-4. Please refer to the tables of the revised manuscript in Page 37-39. The uncertainty analysis was added in Section 3.3.3, please refer to the revised manuscript from Page 27 Line 596 to Page 28 Line 602.

Comment NO.12: *L54: 'important factors' should be 'an important factor'.*

Response: Accepted.

Changes in Manuscript: Please refer to the revised manuscript Page 4 Line 66 for the correction.

Comment NO.13: *L61: 'stationary' should be 'station'.*

Response: Accepted.

Changes in Manuscript: Please refer to the revised manuscript Page 4 Line 75 for the correction.

Comment NO.14: *L64: remove "have been employed".*

Response: Accepted.

Changes in Manuscript: We have rewritten the sentence, please refer to the revised manuscript Page 4 Line 73-77 for the correction.

Comment NO.15: *L64-69: rewrite this sentence. "found that" should have an appropriate subject (e.g., study, human).*

Response: Accepted.

Changes in Manuscript: We rewrote the sentence, please refer to the revised manuscript Page 4 Line 78-80.

Comment NO.16: *L80-84: rewrite this sentence. It is confusing. Do the authors want to say that, when switching from off to on in CMAQ, SO_2 and $PM_{2.5}$ increase by 26% and 15%?*

Response: Accepted.

Changes in Manuscript: We rewrote the sentence, please refer to the revised manuscript from Page 5 Line 94 to Line 98.

Referee #2:

Comment NO.1: *The authors discuss regional air pollutant transport between Beijing and the North China Plain using a 26 1/2 month data set of meteorological parameters and trace gas concentrations. They have a compelling data set and display it with some well chosen plots. However, more rigorous seasonal analysis of the measurements is needed before this paper is publishable.*

Response: Accepted.

Changes in the manuscript: We have added Fig.7 and more seasonal analysis in section 3.2.2 on the seasonal variations of bivariate polar plots for different species. Please refer to the revised manuscript from Page 17 Line 369 to Page 22 Line 474.

Comment NO.2: *Figure 1: To aid the geographically challenged, add topography (mountains), heavily industrialized and urban areas, Gucheng, Baoding-Cangzhou, and Tianjin-Tangshan to Figure 1.*

Response: Accepted.

Changes in Manuscript: Figure 1 was revised following the referee's suggestion.

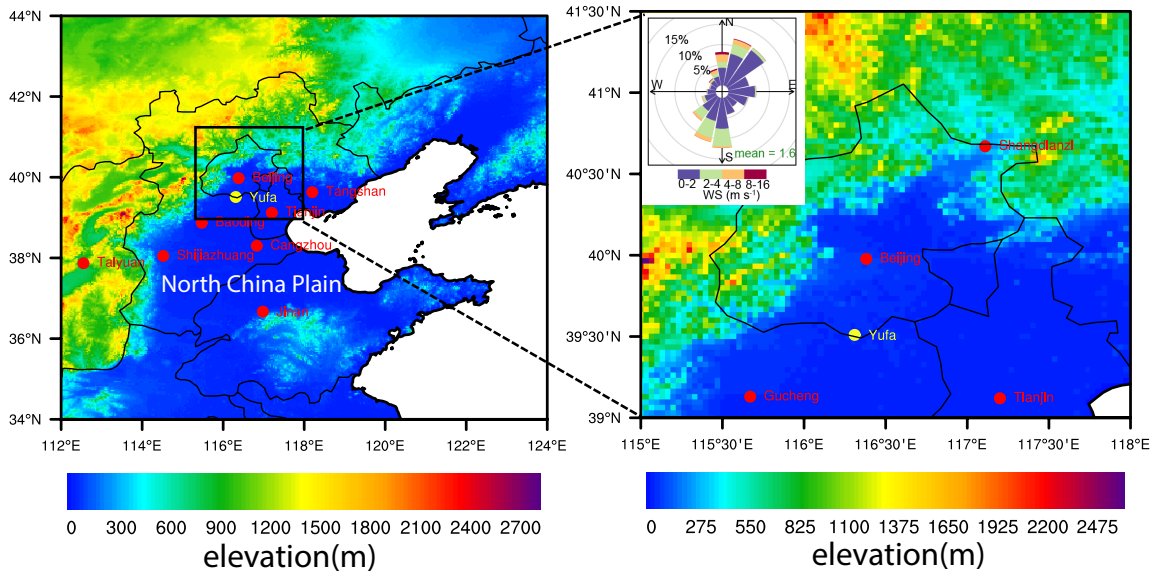

Figure 1. Overview information of the Yufa site.

Comment NO.3: *Figure 2 (Section 3.1) could be replaced by a trace gas version of Figure S2.*

Response: Accepted.

Changes in Manuscript: Please refer to the revised manuscript for Fig.3 in Page 41

Comment NO.4: *Figure 3: In order to strengthen the analysis, you need to separate the data set into seasons. After doing that it will be easier to interpret the results. Trace gas lifetimes vary seasonally and it is unclear what an annually average lifetime means.*

Response: Accepted. We have split the Bivariate Polar plots seasonally for each pollutant in Fig. 7 and added more substantial discussions in the revised manuscript. Moreover, the seasonal differences of the trace gas lifetime were also considered in related discussion in the revised manuscript.

Changes in Manuscript: We have added Fig. 7 in the revised manuscript to strengthen the seasonal analysis. Please refer to the revised manuscript from Page 17 Line 369 to Page 22 Line 474.

Comment NO.5: *I would move S2 and S3 to the main body of the paper.*

Response: Accept.

Changes in Manuscript: We have moved Fig.S2 and Fig.S3 to the main body of the paper. Please refer to the revised manuscript in Page 42-44.

Comment NO.6: *Other Comments: L52: together with → together with a*

Response: Accepted.

Changes in the manuscript: We have rewritten our sentence in the revised manuscript. Please refer to the revised manuscript in Page 4 Line 65.

Comment NO.7: *L83: Models-3/CMAQ model → CMAQ L82/83: up to 26 % and 15 % of what? up to 60 % of what? L89 about 32 %, 11 %, and 3.5 % of what?*

Response: Accepted.

Changes in the manuscript: We have corrected our statement in the revised manuscript and rewrote the sentences. Please refer to the revised manuscript from Page 5 Line 94 to Line 106.

Comment NO.8: *L118: O_x wasn't measured.*

Response: Agree.

Changes in the manuscript: We deleted O_x in this sentence in the revised manuscript. Please refer to the revised manuscript, in Page 7 line 136.

Comment NO.9: *L168-174: This detailed information should be moved to supplemental material and included in the caption for Figure S1.*

Response: Accepted.

Changes in manuscript: We have moved the corresponding content to the supplemental material and included in the caption for Fig. S1.

Comment NO.10: *L180-182: In order to aid in comparison with other studies, give annual means for each trace gas as opposed to the average over a 26 1/2 month period. You can still give the hourly standard deviations but weight them too so that they are not overly influenced by the 2 1/2 month period with 3 years of data.*

Response: Accepted. We have calculated the overall hourly means for each pollutant for the 24 months from from 01 September 2006 to 31 August 2008 to exclude the influence by the data of the 2 1/2 month. The mean±SD (median) concentration value of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO were 15 ± 16 (9) ppb, 12 ± 25 (3) ppb, 24 ± 19 (20) ppb, 36 ± 39 (23) ppb, 28 ± 27 (21) ppb, 52 ± 24 (45) ppb, and 1.6 ± 1.4 (1.2) ppm during the observation period from 01 September 2006 to 31 August 2008, respectively.

Changes in Manuscript: Please refer to the revised manuscript, from Page 13 Line 268 to Line 271 for the changes.

Comment NO.11: *L183/184: observed concentrations of these gaseous pollutants were comparable to reported results This statement is too vague to be of use.*

Response: Accepted. We recalculated the overall hourly means for each pollutant for the two-year observation period and compared our results with the Gucheng site (40.65 °N, 110.11 °E, 293.9 m a.s.l.) and Shangdianzi site (39.13 °N 115.67 °E, 15.2 m a.s.l.), which is one of the regional Global Atmosphere Watch (GAW) stations in China.

The hourly mean±SD (median) concentration value of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO were 15 ± 16 (9) ppb, 12 ± 25 (3) ppb, 24 ± 19 (20) ppb, 36 ± 39 (23) ppb, 28 ± 27 (21) ppb, 52 ± 24 (45) ppb, and 1.6 ± 1.4 (1.2) ppm during the observation period from 01 September 2006 to 31 August 2008, respectively, with hourly mean values -3, 1, 6, 7, -1, 5 and 0 ppb higher for SO_2, NO, NO_2, NO_x, O_3, O_x, and CO than the Gucheng site, a polluted rural site to the south-west of

Beijing, from July 2006 to September 2007 (Lin et al., 2009). The hourly mean values were 12, 11, 17, 28, -5, 22 and 972 ppb higher than those observed at the clean background at the Shangdianzi site, which is one of the regional Global Atmosphere Watch (GAW) stations in China over the period 2004–2006 (Lin et al., 2008). The compared results indicating that Yufa site has become a relatively polluted rural site.

Changes in the Manuscript: We have added detail comparisons with other site, please refer to the revised manuscript from Page 13 Line 268 to Line 279.

Comment NO.12: *L194-196: It should be clarified ...270 to 360 degrees. Move this to the caption for Figure S2. This text detracts from the flow of the manuscript.*

Response: Accepted.

Changes in Manuscript: We have moved the sentences to the caption of the related figure. Please refer to the revised manuscript in Page 42-43.

Comment NO.13: *L205-211: You need to be more rigorous with your analysis here. You can do better than "Generally, the south wind prevailed in the second half of the year ...". and "variations in RH and T were ... typical".*

Response: Accepted. We have added more rigorous analysis in this part.

Changes in Manuscript: We have added more rigorous analysis, please refer to the revised manuscript from Page 14 Line 294 to Line 302.

Comment NO.14: *L207: The seasonal variations L274: Was the reduction in emissions region-wide or mostly in the greater Beijing area?*

Response: The Chinese government has made continuous effort since the 1990s. Moreover, to improve the air quality of 2008 Beijing Olympic Games, the Beijing municipal government implemented strict long- and short-term air pollution control measures including moving heavy polluters out of Beijing city, using low sulfur coal and high standard fuel (e.g. Euro IV) which

made significant decreases in SO_2 in Beijing (Wang et al., 2009, 2011; Qin et al., 2009).

Changes in Manuscript: We have corrected the statement, please refer to the revised manuscript in Page 17 Line 364 to Line 365.

Comment NO.15: *L278-288: Be careful here, yes NO is short-lived but it is also re-generated from NO_2 as part of the NO_x family.*

Response: As previous reported, the conversion efficiency of NO_2 to NO were ~30 % during the period from 15 August to 10 September 2006 (Takegawa et al. 2009). Even when considering the conversion of NO from NO_2 with conversion efficiency ~30 % in summer and autumn (Takegawa et al. 2009), the transport distance of NO is still limited, for the lifetime of NO_2 is also relative short (Beirle et al., 2011; Gu et al., 2013).

Changes in Manuscript: We have considered the transformation of the nitrogen species and added some relevant statement, please refer to the revised manuscript from Page 18 Line 393 to Page 19 Line 396.

Comment NO.16: *L307: The lifetime of SO_2 is not 17 days. Please double check and adjust discussion accordingly.*

Response: Accepted.

Changes in Manuscript: We have corrected SO_2 lifetime in the atmosphere to a couple of hours to 1-2 days, please refer to the revised manuscript from Page 20 Line 431.

Comment NO.17: *L313: essential source → major source L323: Beijing area closely → Beijing area are closely L329: transport of SO_2, CO_2, ... were → transport of SO_2, CO_2, ... was L344: the transport direction → the net transport direction*

Response: Accepted.

Changes in Manuscript: We have made relevant revisions following the referee's suggestion,

please refer to the revised manuscript in Page 20 Line 438, Page 21 Line 462, and Page 22 Line 481.

Comment NO.18: *L384: You need to add more detail here rather than stating that fluxes were lower in 2008 than 2007. For example, the decrease in summertime CO and NOx fluxes between 2007 and 2008 is on the order of a factor of 5.*

Response: Accepted.

Changes in Manuscript: We have added more discussions in this part, please refer to the revised manuscript from Page 24 Line 515-519.

Comment NO.19: *L401-404: You need to convince the reader that the use of surface winds and trace gas concentrations at one site is sufficient to calculate the flux of a trace gas between the two regions.*

Response: Considering the limitation of the spatial representation of the site and lack of the 3 dimensional observation data, we have made major revision about this study and focus on the regional transport influence of Beijing and the NCP at this cross-boundary site, the Yufa site.

Changes in Manuscript: Please refer to the revised manuscript in the Abstract from Page 2 Line 19 to Page 3 Line 54 and Conclusions from Page 28 Line 613 to Page 30 Line 650.

Comment NO.20: *L417: The conclusion section is too similar to the abstract. Expand it a bit. Figure S2 caption for BP should be hPa.*

Response: Accepted.

Changes in Manuscript: We have expanded the conclusion section based on the major revision of the manuscript. Please refer to the revised manuscript from Page 28 Line 613 to Page 30 Line 650. The mistake in Fig. 3 caption for BP was also corrected in the revised manuscript, please refer to the revised manuscript in Page 42.

Comment NO.21: *In order to aid the reviewer, please order the wind rose plots as follows (you can then compare similar seasons easily). a: Autumn 2006 b: Autumn 2007 c: Winter 2006/07 d: Winter 2007/08 e: Spring 2007 f: Spring 2008 g: Summer 2007 h: Summer 2008*

Response: Accepted.

Changes in Manuscript: We have made the corresponding adjustment of the related figure. Please refer to the revised manuscript in Page 44.

Comment NO.22: *Domain for Figure S4 should match that for Figure 1. Add location of Beijing.*

Response: Accepted.

Changes in Manuscript: We have made revision of Fig. 1 and Fig. S3 in the revised manuscript as the referee's suggestion, please refer to the revised manuscript in Page 40 and Page 46.

Referee #3:

Comment NO.1: *The authors showed the observations of short-lived gases (SO_2, NO, NO_2, NO_x, O_3, O_x, and CO) during two years at the Yufa site where is located between Beijing and the North China Plain (NCP). The observation for such long-term period is limited in China, so that the results are important for understanding the real situation of the air pollutions. They also analyzed fluxes of the air pollutions at the Yufa site using observed winds and a simple flux analysis. It is a pity that the authors do not use any models including a back trajectory to analyze the fluxes. In the manuscript, some of important points are missed, but in overall the manuscript would be acceptable for publication if these comments can be satisfactorily addressed.*

Response: Accepted. We have made major revision in our paper and added PSCF analysis based on the back trajectory model (the HYSPLIT model) to give more convincing validations of our results.

Changes in Manuscript: The PSCF analysis based on HYSPLIT model was added to back our

results of regional transport influence of Beijing and the NCP on the Yufa site. Please refer to the revised manuscript in section 2.2.3 from Page 11 Line 232 to Page 12 Line 262, section 3.3.2 from Page 26 Line 567 to Page 27 Line 595, and also Fig. 9 and 10 in Page 49-50.

Comment NO.2: *L49-50: The location of NCP is unclear for me and probably most readers. Please add the exact location as well as the actual topology in Figure 1.*

Response: Accepted.

Changes in Manuscript: Figure 1 was revised following the referee's suggestion.

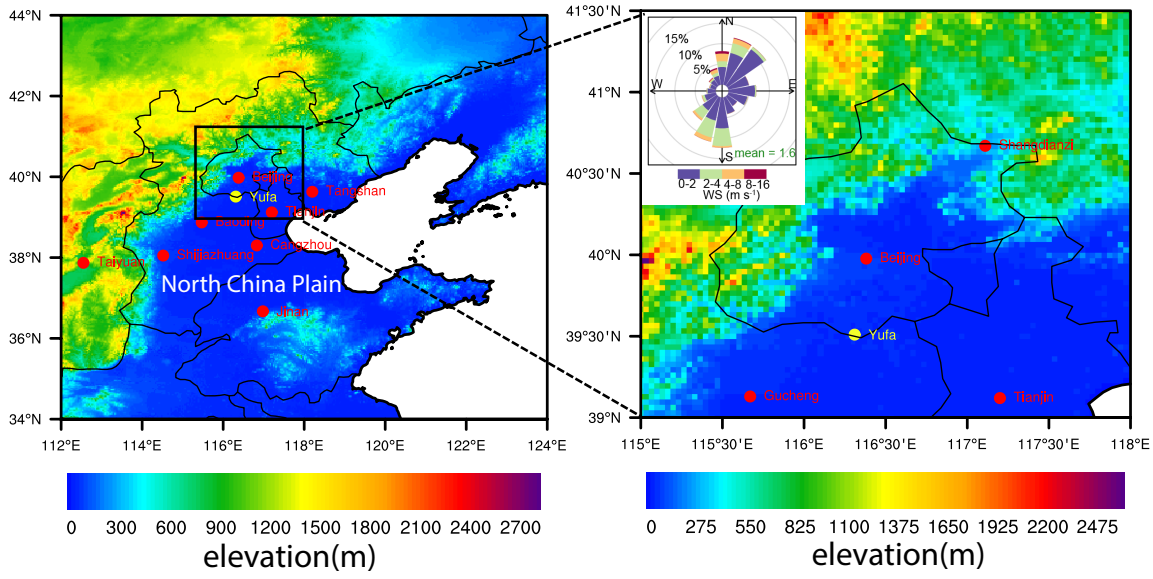

Figure 1. Overview information of the Yufa site.

Comment NO.3: *L118-L128: What is the instrument uncertainty in this study? That means the accuracy of each instrument must be shown here.*

Response: Accepted. We have added more detailed information of the instrument in the revised manuscript and listed it in Table 1.

Table 1. The information of the measurement instruments.

Species/ Parameter	Instrument	Detection limit	Time resolution	Precision	Uncertainty
SO_2	Ecotech 9850B	0.3 ppb	1 min	0.5% (1ppb)	5%
$NO\text{-}NO_x$	Ecotech 9841B	50 ppb	1 min	1% (0.5ppb)	10%
CO	Ecotech 9830	0.4 ppb	1 min	1% (0.1ppm)	1%
O_3	Ecotech 9810B	0.5 ppb	1 min	0.5% (1ppb)	5%
WS	LASTEM	-	10 min	0,1 m s^{-1}	5%
WD	LASTEM	-	10 min	0,1 º	1%
BP	LASTEM	-	10 min	0,1 hPa	±0.35 hPa
T	LASTEM	-	10 min	0,1 ℃	±0.2°C
RH	LASTEM	-	10 min	1%	±3%

Changes in Manuscript: We have added more detailed information of the precisions and uncertainties of the measurement, please refer to the revised manuscript, from Page 7 Line 136 to Page 8 Line 158.

Comment NO.4: *L192: Figure S2 must be moved to the manuscript (NOT supplement).*

Response: Accepted.

Changes in Manuscript: We have moved Fig. S2 to the main text as Fig. 3, please refer to the revised manuscript in Page 42-43.

Comment NO.5: *L204: Figure S3 is also an important figure in your manuscript, so please move it to the manuscript (NOT supplement).*

Response: Accepted.

Changes in Manuscript: We have moved Fig. S3 to the main text as Fig. 4, please refer to the revised manuscript in Page 44.

Comment NO.6: *L314-322: Unlike the other species, O_3 is a secondary product. The O_3 flux change is not so simple that the analysis only using observed winds and prescribed emission inventories may not be enough. The discussion in the annual mean values is also rough, since the seasonal variation of O_3 distribution is large due to the seasonality of the meteorological fields, the height of the boundary layer, and O_3 chemistry. At least, a seasonal analysis is*

required.

Response: Agree.

Changes in Manuscript: We have added more seasonal analysis of O_3 in the revised manuscript following the referee's suggestion. Please refer to the revised manuscript from Page 21 Line 442-461.

Comment NO.7: *L363-364: The authors mention that the fluxes of the pollutions in winter of 2006 are unusual, but there is measurement only two years (2006 and 2007). How do the authors determine the specialty of 2006? Please clarify it.*

Response: Agree.

Changes in Manuscript: We have corrected our statement, please refer to the revised manuscript from Page 24 Line 515-519.

Comment NO.8: *L372-373: It seems to me that Figure S4 suggests the strong peaks are found over NCP as well as Beijing. What is the evidence of the statement "partly attributed to the high emission intensity of SO_2 in the NCP and the reduction of SO_2 emission in Beijing"?*

Response: We have given the spatial distribution of seasonal NO_x, CO, and SO_2 emissions in Northern China in 2008 based on the Multi-resolution Emission Inventory of China (MEIC; www.meicmodel.org) in Fig. 6 in the revised manuscript. And Fig. 6 showed the emission intensity of SO_2 in the NCP is somewhat higher than that in Beijing in spring, summer and autumn. Moreover, to improve the air quality of 2008 Beijing Olympic Games, the Beijing municipal government implemented strict long- and short-term air pollution control measures including moving heavy polluters out of Beijing city, using low sulfur coal and high standard fuel (e.g. Euro IV) which made significant decreases in SO_2 in Beijing (Wang et al., 2009, 2011; Qin et al., 2009).

Changes in manuscript: We have added Fig. 6, please refer to the revised manuscript in Page 46,

and rewrote the relevant sentence in Page 17 Line 364-365.

Comment NO.9: *L379-382: This discussion is very important but too shallow. The discussion for the difference in the fluxes of the pollutant between 2008 and the other year strongly supports the author's conclusion shown in L41-42. Please add more discussion here.*

Response: Agree. We have added more in-depth discussion in the manuscript for the Olympic Game period. And the mean surface flux intensity and concentrations of pollutants for the period of 08.08.2007-24.08.2007 and 08.08.2008-24.08.2008 were calculated and compared and listed in Table 4.

Table 4. The mean surface transport flux intensity (i.e. Flux_2007), the influx intensity (i.e. the positive surface flux, from the NCP to Yufa, In_2007 and In_2008), the outflux intensity (i.e. the negative surface flux, from Beijing to Yufa, Out_2007 and Out_2008), and the mean concentration during the 2008 Beijing Olympic period (from 8 August 2008 to 20 September 2008) and the same corresponding period of 2007 (from 8 August 2007 to 20 September 2007).

Flux ($\mu g\ s^{-1}\ m^{-2}$)	Flux_2007	In_2007	Out_2007	Flux_2008	In_2008	Out_2008	Cont._2007 (ppb)	Cont._2008 (ppb)
SO_2	7.9±19.3	14.9±20.8	-4.5±4.6	1.4±15.5	11.9±13.6	-9±8.8	3.6±3.4	3.9±2.2
NO	0.3±8.6	3.8±5	-5.9±9.9	-0.1±3.2	2.4±1.8	-2.5±2.3	4.3±5.5	1.9±0.6
NO_2	4.1±37.9	24.1±18.6	-31.3±37.9	-1.3±21.9	15.2±11.6	-17.5±17.1	16.1±10.2	8.5±3.6
NO_x	4.4±44.5	27.8±20.7	-37.2±45.3	-1.4±25	17.5±13.2	-20±19.2	20.5±13.3	10.4±4
CO	540±158	1390±1160	-980±980	10±1110	870±670	-850±740	1190±490	750±260
O_3	60±130	117.9±122.6	-42.6±61.1	24.9±124.6	110.9±111	-60.6±63.7	41.1±30.5	38.9±25.8
O_x	64.1±154.4	141.9±129.1	-73.9±82.4	23.7±142.1	126.7±118.4	-77.7±74.6	57.2±27.3	47.4±24.1

Changes in Manuscript: Please refer to the revised manuscript in Page 39 for Table 4 and from Page 25 Line 535 to Line 549 for the relevant discussions.

Comment NO.10: *L385-391: In general, the spatial distribution of O_3 tends to be broader (non-localized) than that of primary species such as NO_x and CO. So, I suppose the difference between O_3 and NO_x is mainly caused by the difference between primary and secondary sources. I don't understand the short distance is the primary reason of the difference. In addition, what do the*

authors mean "underestimation" in L388 and "overestimate" in L390? Please explain it.

Response: Accepted.

Changes in Manuscript: We have added in-depth discussion on ozone and deleted the misleading statement in the revised manuscript.

Comment NO.11: *L398-400: I understand the flux calculation used in this study includes various limitation, but more discussion for the uncertainty of the method is required.*

Response: Accepted. Uncertainty in calculation of the surface flux intensities in this study mainly come from the measurement of the pollutants and the wind. The uncertainty of the measurement for SO_2, NO_x, CO, and O_3 is within 10 %, 10 %, 1 %, and 5 %, respectively. The uncertainty of wind speed is less than 5 % and the uncertainty of wind direction is about 1 %. Thus the overall surface flux strength calculation uncertainty for SO_2, NO_x, CO, and O_3 is less than 12 %, 12 %, 6 % and 8 %.

Changes in Manuscript: We have added discussion on the uncertainty of the flux calculation in the revised manuscript in Section 3.3.3, please refer to the revised manuscript from Page 27 Line 596 to Page 28 Line 602.

Comment NO.12: *L405-408: I don't understand what the authors want to explain. Why is it impossible to apply the method to the other sites? Please clarify it. This is related to my comment #10.*

Response: We have deleted the misleading sentences in the revised manuscript.

Changes in Manuscript: We corrected our statement, please refer to the revised manuscript from Page 26 Line 560 to Line 566.

Comment 13: *L414: The topography around Beijing is unknown among general readers, so please show the topography in Figure. This is related to my comment #1.*

Response: Accepted.

Changes in Manuscript: We have added the topography around Beijing in Fig. 1, please refer to the revised manuscript in Page 40.

Comment NO.14: *Abstract: I recommend the authors also show the observed concentrations of gases in annual means at the Yufa site, because the two-year observation in China become an important information.*

Response: Accepted. As the referee's suggestion, we put the sentence "The hourly mean±SD (median) concentration value of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO were 15 ± 16 (9) ppb, 12 ± 25 (3) ppb, 24 ± 19 (20) ppb, 36 ± 39 (23) ppb, 28 ± 27 (21) ppb, 52 ± 24 (45) ppb, and 1.6 ± 1.4 (1.2) ppm during the observation period from 01 September 2006 to 31 August 2008, respectively." to the abstract in the revised manuscript.

Changes in Manuscript: We have added the sentence to abstract, please refer to the revised manuscript in Page 2 Line 30 to Line 33.

Comment NO.15: *L36-38: More details of the quantitative values would be preferred here.*

Response: Accepted. We have given more quantitative results in the abstract in the revised manuscript. "The surface transport flux intensities (mean ± SD) of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO were 6.2±89.5, -4.3±29.5, -0.6±72.3, -4.9±93, 14.7±187.8, 14.8±234.9, and 70±2830 μg s^{-1} m^{-2} during the observation period, respectively. For SO_2, CO, O_3, and O_x the surface flux intensities from the NCP to Yufa surpassed those from Beijing to Yufa in all seasons except winter, with the strongest net fluxes largely in summer, which is about 4–8 times of other seasons. The surface transport flux intensity of NO_x from Beijing to Yufa was stronger than that from NCP to Yufa except in summer, with the strongest net flux in winter, which is about 1.3–8 times of other seasons."

Changes in Manuscript: Please refer to the revised manuscript in Page 2 Line 37 to Line 45.

Comment NO.16: *L180-L184: How about the air quality level at the Yufa site compared to the other sites in China and out of China like megacities in Asia, US, and Europe? Although the author mention "comparable to reported results at Gucheng site", the authors can add actual values at Gucheng and other sites using at least results in literatures referred in section 1.*

Response: Accepted. We recalculated the overall hourly means for each pollutant for the two-year observation period and compared our results with the Gucheng site (40.65 °N, 110.11 °E, 293.9 m a.s.l.) and Shangdianzi site (39.13 °N 115.67 °E, 15.2 m a.s.l.), which is one of the regional Global Atmosphere Watch (GAW) stations in China.

The hourly mean±SD (median) concentration value of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO were 15 ± 16 (9) ppb, 12 ± 25 (3) ppb, 24 ± 19 (20) ppb, 36 ± 39 (23) ppb, 28 ± 27 (21) ppb, 52 ± 24 (45) ppb, and 1.6 ± 1.4 (1.2) ppm during the observation period from 01 September 2006 to 31 August 2008, respectively, with hourly mean values -3, 1, 6, 7, -1, 5 and 0 ppb higher for SO_2, NO, NO_2, NO_x, O_3, O_x, and CO than the Gucheng site, a polluted rural site to the south-west of Beijing, from July 2006 to September 2007 (Lin et al., 2009). The hourly mean values were 12, 11, 17, 28, -5, 22 and 972 ppb higher than those observed at the clean background at the Shangdianzi site, which is one of the regional Global Atmosphere Watch (GAW) stations in China over the period 2004–2006 (Lin et al., 2008). The compared results indicating that Yufa site has become a relatively polluted rural site.

Changes in the Manuscript: We have added detail comparisons with other site, please refer to the revised manuscript from Page 13 Line 268 to Line 279.

Comment NO.17: L205: *What is the definition of the four seasons in your manuscript? Does the winter represent DJF? Please clarify it.*

Response: Accepted. In our study, spring included March, April, and May (MAM); summer included June, July, and August (JJA); fall Included September, October, and November (SON); winter included December, January and February (DJF).

Changes in Manuscript: We have given the clarification of seasons in the right place, please refer to the revised manuscript in Page 44.

Comment NO.18: L306-307*: The atmospheric lifetime of SO_2 must be much shorter than 17 days. It is probably several days.*

Response: Accepted.

Changes in Manuscript: We have corrected SO_2 lifetime in the atmosphere to a couple of hours to 1-2 days, please refer to the revised manuscript from Page 20 Line 431.

Comment NO.19: L341-342: *Please compare these values with those obtained by other sites in the world?*

Response: As the surface flux intensity calculations based on the long-term station observation is still lack up to now, it is difficult to compare our results with other sites in the world. However, our results somewhat agreed with the results of previous research that the influx of SO_2 to the megacity Beijing from the surrounding areas in the south (An et al., 2007; Wang et al., 2011).

Referee #4

Comments NO.1: *This work presents an interesting study on the regional-flux calculation based on two- year valuable ground-based measurement at a cross-boundary site between Beijing and its neighbor province. Generally speaking, the overall scientific topic of this paper, i.e. the cross-boundary transport, is very important for air quality management. The authors tried to apply a method to estimate the pollution flux based on ground-based measurement following the idea proposed 40 years ago by White et al. (1976).*

In White's work, aircraft and balloon measurements in different locations in and down- wind a city were used for the flux estimation, here this study was mainly based ground based measurements. In air quality management, the flux around surface is mean- ingless but a flux in the entire boundary layer is the main concern. However, because the cross-boundary PBL

transport flux at a certain place will be strongly related to the distribution of vertical profiles of air pollutants and wind. Primary pollutants like SO_2, CO, NO_x generally have a different vertical profile with secondary species like O_3, especially at nighttime. So the flux calculation based on ground-based data will have very large uncertainty at different time of a day.

This paper focuses more on method application/developing but not results discussion, which maybe the reason that they only put several key figures in the main text but others in the supplementary. For this reason, although the authors already mentioned that they didn't consider the possible influence from high altitude and boundary layer (Line 402-404), I would like to encourage them pay more efforts to improve the methods of PBL flux calculation based on ground-based station measurement. Below I suggest some possible methods for this kind of calculation.

First, because nowadays numerical models, such as the CMAQ or WRF-Chem, could give a relatively good estimation in the transport flux at surface and also the entire PBL, I would like suggest the authors to make a comparison of their calculated flux with these modeling results. This kind of modeling study and comparison don't have to cover very long period but may be okay for several days in different seasons. This comparison will provide valuable information on how to use the ground-based data to estimate the PBL flux. One interesting point could be that if the proposed method only works for the early afternoon (12:00-16:00 for example), when the boundary layer is well-developed and the vertical profiles of air pollutants and wind are relatively unified in the entire boundary layer. If it is true, maybe the authors can only use the afternoon data to discuss the seasonal pattern of flux.

If the authors would like to further apply this method for other time of a day except the early afternoon, they could use historical averaged profiles of air pollutants and wind to give an estimation of the PBL flux. In fact, in Beijing and other regions in the North China Plain some studies already showed vertical profiles of air pollutants based on aircraft or balloon studies and also there are routine radiosonde measurements of wind in some meteorological station. It will be better to include those averaged profiles in the estimation of PBL flux based on surface

measurement. Of course, these modeling studies for cases in different seasons will be useful for evaluating these calculations and for improving the methodology.

Response: The critical comments and suggestions of the reviewer are highly appreciated. We agree with the reviewer that the cross-boundary transport is very important for air quality management, and we are developing a method to partly fulfil this purpose. Due to the lack of vertical profiles of wind speed and air pollutant concentrations, in our revised manuscript, we focus on the surface flux intensity calculation based on the two-year ground measurement at a cross-boundary site between Beijing and the NCP to investigate the surface regional transport influence of Beijing and the NCP at the Yufa site, and we do not intend to extrapolate the results into the whole boundary layer height, based on the following consideration:

(1). The novelty of our study is to develop a method based on long-term ground based measurement to calculate the surface transport flux intensity across a cross-boundary site. We need to make it clear that the surface flux intensity calculated in this study is the per unit area flux across the Yufa site, which is different from the flux across a large area reported in other studies (e.g. Wang et al. 2011). Our results could only be extrapolated if all the concentrations of all the pollutants and wind speed and direction were homogenously distributed, vertically and horizontally. Otherwise, we need vertical profiles of air pollutants concentration and wind to calculate the cross-section transport flux of two adjacent regions for the whole boundary layer by the integrating formula: $FLUX = \iint C_{(x,z)} WS_{(x,z)} sin\theta_{(x,z)} dxdz = \iint f_{(x,z)} dxdz$; where x is horizontal distance to the observed point, z is the vertical distance from ground to the observed point.

In this study, we did not intent to extrapolate from Yufa site to the entire region. We only focus on the method developing and evaluation of the regional transport influence of Beijing and the NCP on the cross-boundary site based on the ground-based observation data. We conducted Bivariate Polar plots analysis and surface flux intensity calculation and get evidence of surface pollutants transport from Beijing to the Yufa site and from the NCP to the Yufa site. Considering the variations of the vertical and horizontal distributions of the air pollutants and

meteorological parameters, and influence of the boundary layer on the regional transport, more multidimensional data with high precision and resolution are needs for further comprehensive discussion of the regional transport between Beijing and NCP.

(2). We are also aware the limitation of the spatial representation of the Yufa site, so we added the back trajectory model to validate our results. As a cross-boundary site between the megacity Beijing and the NCP, the surface transport flux strengths at the Yufa site may also indicate the transport between the megacity Beijing and the NCP.

(3). The referee's two suggestions on improving the methods of PBL flux calculation based on ground-based station measurement through model validation and historical data extrapolation are very valuable and highly appreciated. However, we think it is better conducted in a separate study and published in a separated paper. Otherwise we will lose the focus of our methodology developing. Besides, it is difficult for us to obtain the history profile data with high precision and resolution during the observation period of Yufa, so the extrapolation base on the historical profiles may contain large errors.

Comments NO.2: *Minor comments: 1) Figure 1, it will be better to include terrain and also the emission data in the figure.*

2) White et al. (1976), one of the most important references, was missed in the reference list.

Response: Accepted.

Changes in Manuscript: We have added the topography around Beijing in Fig. 1, please refer to the revised manuscript in Page 40; the missing citation for White et al. was added, please refer to the revised manuscript, from Page 35 Line 784 to Line 786.

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