Response to the Comments of Referees

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

Yingruo Li, Chunxiang Ye, Jun Liu, Yi Zhu, Junxia Wang, Ziqiang Tan, Weili Lin, Limin Zeng, Tong Zhu

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, adding 6 more figures and in-depth discussion in the main text as suggested.

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 49-52; we also added more

discussions relevant to the figures, please refer to the revised manuscript Section 3.2.2 from Page 17 Line 367 to Page 22 Line 472 for the discussions to Fig.6-7 and Section 3.4 from Page 26 Line 568 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 uncertainties 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 192 to Page 11 Line 229.

We have added more detailed information of the precisions and uncertainties of the measurement, please refer to the revised manuscript, from Page 7 Line 134 to Page 8 Line 155.

The uncertainty analysis was added in Section 3.5, please refer to the revised manuscript from Page 27 Line 596 to Page 28 Line 614.

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 230 to Page 12 Line 260, section 3.4 from Page 26 Line 568 to Page 27 Line 595, and also Fig. 9 and 10 in Page 49-52.

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: 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 vector 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 its 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 of 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 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 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 obtained clear evidences 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, three dimensional data with high precision and resolution are needed 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) in the campus of Huangpu College. There is no tall building around the Yufa site which affects the wind and gaseous pollutant 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 192 to Page 11 Line 229; 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 119 to Line 132; the discussion relevant to (4) The extrapolation from Yufa site to the entire region, please refer to the revised manuscript from Page 27 Line 596 to Line 614.

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 67 and Page 5 Line 88.

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 78 to Line 80.

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.

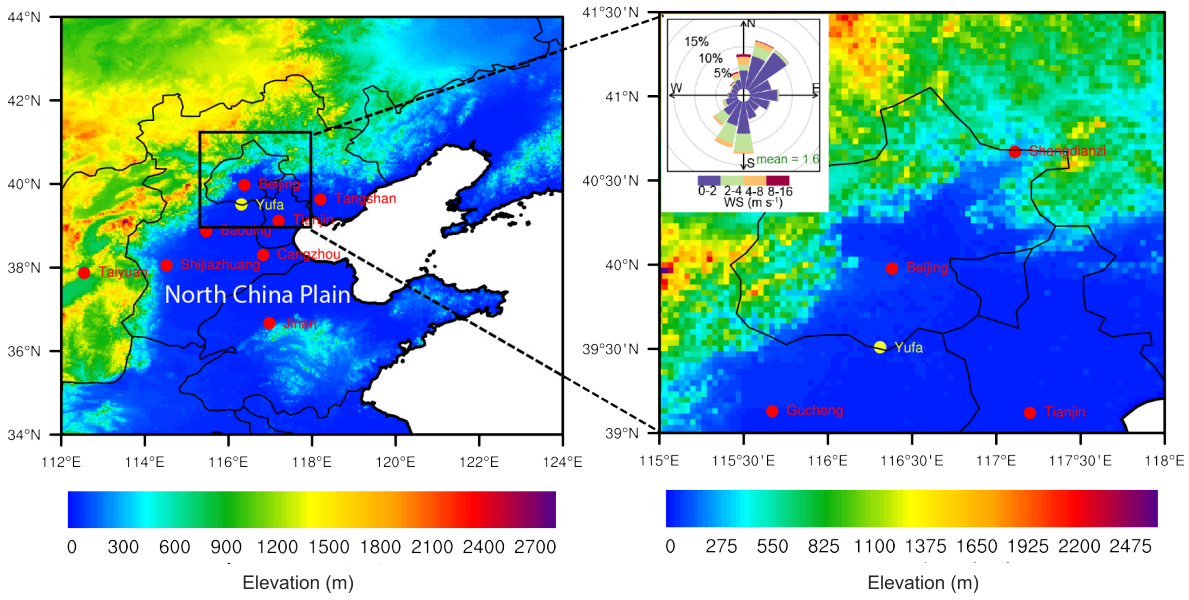

Figure 1. Overview information of the Yufa site.

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	0.5 ppb	1 min	0.5% (0.5 ppb)	10%
NO-NO_x	Ecotech 9841B	0.5 ppb	1 min	1% (0.5 ppb)	10%
CO	Ecotech 9830	50 ppb	1 min	1% (0.1 ppm)	1%
O_3	Ecotech 9810B	0.4 ppb	1 min	0.5% (1 ppb)	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 134 to Page 8 Line 155; the missing citation for White et al. was added, please refer to the revised manuscript, from Page 35 Line 783 to Line 785; 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 150-151. 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 185 to Line 191.

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 367 to Page 22 Line 472.

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 in Page 20 Line 428.

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 3.5, please refer to the

revised manuscript from Page 27 Line 596 to Page 28 Line 614.

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

Response: Accepted.

Changes in Manuscript: Please refer to the revised manuscript in Page 4 Line 64 for the correction.

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

Response: Accepted.

Changes in Manuscript: Please refer to the revised manuscript in Page 4 Line 73 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 in Page 4 Line 71-75 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 in Page 4 Line 76-81.

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 in Page 5 Line 92-96.

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 367 to Page 22 Line 472.

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.

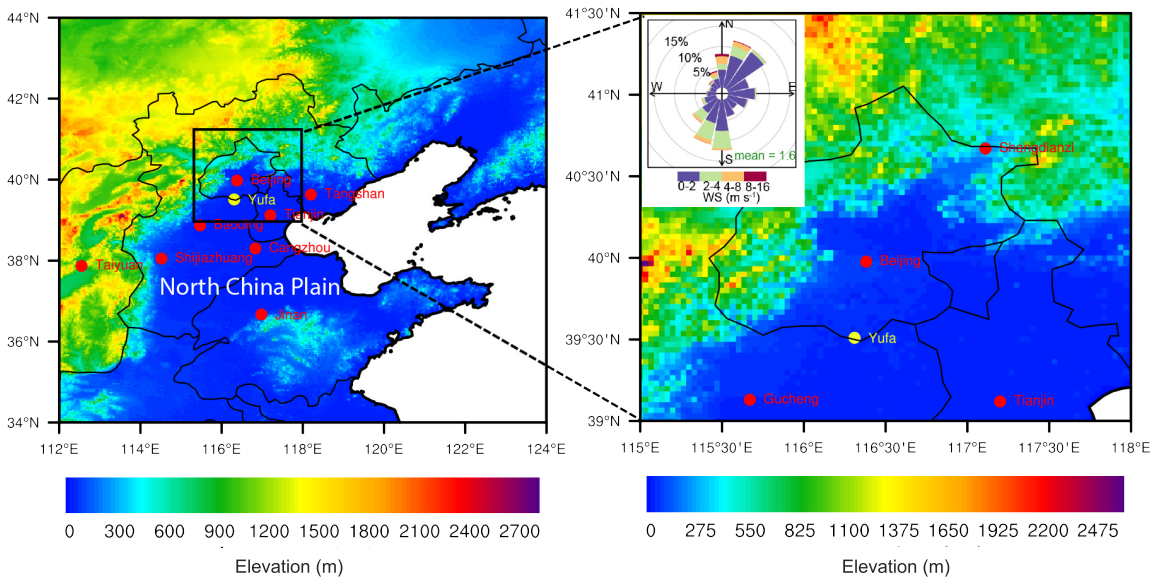

Figure 1. Overview information of the Yufa site.

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 42-43.

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 367 to Page 22 Line 472.

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 as Fig.3 and Fig.4. 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 63.

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 in Page 5 Line 98-104.

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 134.

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 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.

Changes in Manuscript: Please refer to the revised manuscript, from Page 13 Line 266 to Line 270 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.), a rural site to the south-west of Beijing, 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 266 to Line 277.

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 293 to Line 300.

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 (Qin et al., 2009; Wang et al., 2009, 2011).

Changes in Manuscript: We have corrected the statement, please refer to the

revised manuscript in Page 17 Line 362 to Line 363.

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 was ~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 391 to Page 19 Line 394.

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 428.

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 435, Page 21 Line 460, and Page 22 Line 477.

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 516-518.

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 three 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 18 to Page 3 Line 52 and Conclusions from Page 28 Line 616 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 616 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 for Fig.4 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 230 to Page 12 Line 260, section 3.4 from Page 26 Line 568 to Page 27 Line 595, and also Fig. 9 and 10 in Page 49-52.

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.

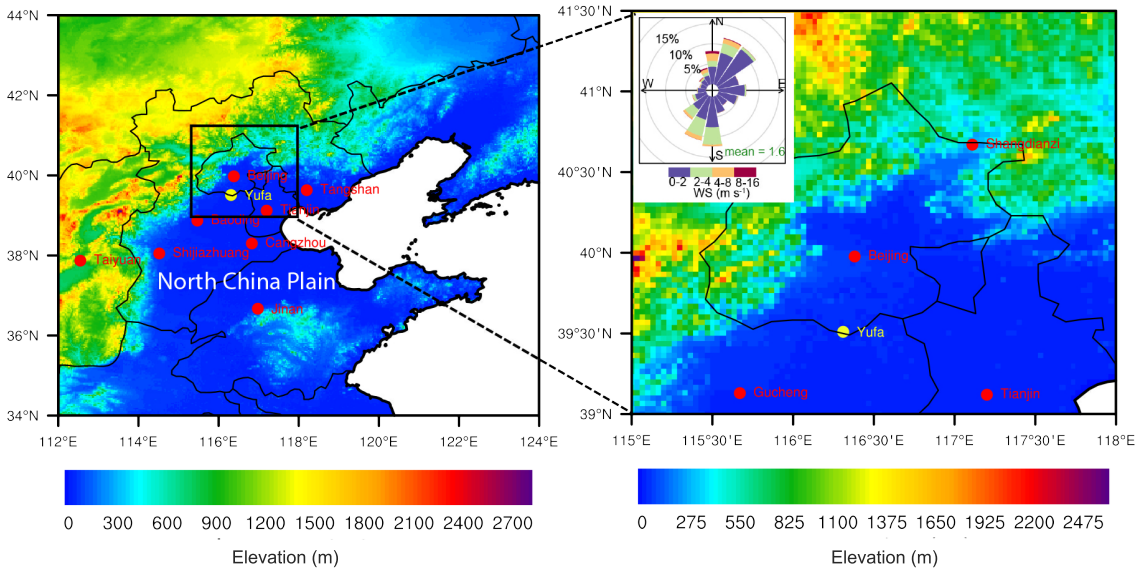

Figure 1. Overview information of the Yufa site.

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	0.5 ppb	1 min	0.5% (0.5 ppb)	10%
NO-NO_x	Ecotech 9841B	0.5 ppb	1 min	1% (0.5 ppb)	10%
CO	Ecotech 9830	50 ppb	1 min	1% (0.1 ppm)	1%
O_3	Ecotech 9810B	0.4 ppb	1 min	0.5% (1 ppb)	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 134 to Page 8 Line 155.

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 in Page 21 Line 440-459.

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 in Page 24 Line 513-518.

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 (Qin et al., 2009; Wang et al., 2009, 2011).

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 362-363.

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 net surface transport flux intensities (i.e. Flux_2007 and Flux_2008), the influx intensities (positive; from the NCP to Yufa; In_2007 and In_2008), the outflux intensities (negative; from Beijing to Yufa; Out_2007 and Out_2008), and the mean concentrations (i.e. Cont._2007 and Cont._2008) 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 534 to Line 548 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 comes from the measurement of the pollutants and the wind. Based on the instruments used, the uncertainty of the measurement of the concentrations of SO_2, NO_x, CO, and O_3 was within 10 %, 10 %, 1 %, and 5 %, respectively. The uncertainty of wind speed measurement was less than 5 % and the uncertainty of wind direction was about 1 %. Thus, the uncertainty of the overall surface flux intensity for SO_2, NO_x, CO, and O_3 was less than 12 %, 12 %, 6 %, and 8 %, respectively.

Changes in Manuscript: We have added discussion on the uncertainty of the flux calculation in the revised manuscript in Section 3.5, please refer to the revised manuscript from Page 27 Line 596 to Page 28 Line 603.

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 561 to Line 567.

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 29 to Line 32.

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 net surface transport flux intensity (mean ± SD) of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO was 6.2 ± 89.5, -4.3 ± 29.5, -0.6 ± 72.3, -4.9 ± 93.0, 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 wint er, with the strongest net fluxes largely in summer, which was about 4–8 times of other seasons. The surface transport flux intensity of NO_x from Beijing to Yufa was stronger than that from the NCP to Yufa except in summer, with the strongest net flux in winter, which was 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.), a rural site to the south-west of Beijing, 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 was 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 indicated that the 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 266 to Line 278.

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 428.

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 continuous 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 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 obtained clear evidences 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, three dimensional data with high precision and resolution are needed 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 783 to Line 785.

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

Yingruo Li[1], Chunxiang Ye[1,2], Jun Liu[1], Yi Zhu[1], Junxia Wang[1], Ziqiang Tan[1], Weili Lin[3], Limin Zeng[1], Tong Zhu[1*]

[1]SKL-ESPC and BIC-ESAT, College of Environmental Sciences and Engineering, Peking University, Beijing, 100871, China

[2]Now at School of Chemistry, University of Leeds, Leeds LS2 9JT, UK

[3]Meteorological Observation Center, China Meteorological Administration, Beijing, 100081, China

*Corresponding Author: tzhu@pku.edu.cn

Abstract. Megacities have strong interactions with the surrounding regions through transport of air pollutants. It has been frequently addressed that the air quality of Beijing is influenced by the influx of air pollutants from the North China Plain (NCP). Estimations of air pollutant cross-boundary transport between Beijing and the NCP are important for air quality management. However, evaluation of cross-boundary transport using long-term observations is very limited. Using the observational results of the gaseous pollutants SO_2, NO, NO_2, O_3, and CO from August 2006 to October 2008 at the Yufa site, a cross-boundary site between the megacity Beijing and the NCP, together with meteorological parameters, we explored a method for evaluating the transport flux intensities at Yufa, as part of the "Campaign of Air Quality Research in Beijing and Surrounding Region 2006–2008" (CAREBeijing 2006–2008). The hourly mean ± SD (median) concentration of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO was 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, respectively. The bivariate polar plots showed the dependence of pollutant concentrations on both wind speed and wind direction, and thus inferred their dominant transport directions. Surface flux intensity calculations further demonstrated the regional transport influence of Beijing and the NCP on Yufa. The net surface transport flux intensity (mean ± SD) of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO was 6.2 ± 89.5, -4.3 ± 29.5, -0.6 ± 72.3, -4.9 ± 93.0, 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

was about 4–8 times of other seasons. The surface transport flux intensity of NO_x from Beijing to Yufa was stronger than that from the NCP to Yufa except in summer, with the strongest net flux in winter, which was about 1.3–8 times of other seasons. The flux intensities were then assigned to the corresponding trajectories in the potential source contribution function analysis (PSCF), which confirmed the results of flux intensity calculations. Our study also suggested that various factors, such as the wind field, emission inventory, and photochemical reactions, could influence transport of air pollutants. The decrease of surface flux intensity during the Olympic Games period implied the role of both local emission reduction and regional cooperation in successful air quality management. Three dimensional observations are needed for further comprehensive discussion of the regional transport between Beijing and the NCP.

Keywords: Megacity Beijing, North China Plain, Yufa site, Regional transport, Long-term and multiple-species observation

1. Introduction

Megacities are large sources of air pollutants and greatly influence the surrounding areas (Parrish and Zhu, 2009). With a population over 20 million, the city of Beijing is an example of such a megacity. Beijing has faced severe air pollution problems over the past two decades and has intensive interactions with other emission hot spots within the North China Plain (NCP) (Chen et al., 2015; Shao et al., 2006; Zhang et al., 2012). Beijing and the NCP are surrounded by the Yanshan Mountains to the north and the Taihang Mountains to the west. The semi-basin geographical features

63 together with the continental monsoon climate make regional transport of air
64 pollutants between the megacity Beijing and the NCP an important factor affecting
65 air quality in Beijing and the NCP (An et al., 2007; Guo et al., 2010; Lin et al., 2008,
66 2009; Streets et al., 2007; Wang et al., 2006; Wang et al., 2011; Wang et al. 2015; Wu
67 et al., 2011; Xu et al., 2005; Xu et al., 2011). An improved understanding of the
68 regional transport of air pollutants between Beijing and the NCP is therefore
69 essential for air quality management of the megacity Beijing and establishment of
70 regional-scale emissions control measures.

71 Previous studies have shed light on the regional transport sources of the
72 megacity Beijing, and various techniques have been employed, including rural/urban
73 station observations (Guo et al., 2010; Lin et al., 2008, 2009; Wang et al., 2006; Xu et
74 al., 2011), mobile laboratory measurements (Wang et al., 2009, 2011; Zhu et al.,
75 2016), and modelling studies (An et al., 2007; Matsui et al., 2009; Wu et al., 2011). A
76 ground-based observation study from July 2006 to September 2007 at the Gucheng
77 site (Lin et al., 2009), a rural site south-west of Beijing, found that high
78 concentrations of gaseous pollutants, including nitric oxide (NO), nitrogen dioxide
79 (NO_2), nitrogen oxides (NO_x=NO+NO_2), sulphur dioxide (SO_2), carbon monoxide (CO),
80 ozone (O_3), and oxidant (O_x=NO_2+O_3), were accompanied by air masses moving
81 northward from Gucheng to Beijing, according to back-trajectory analysis. Similar to
82 Lin et al. (2009), regional transport of air pollutants between Beijing and the NCP was
83 observed consistently in these previous studies (Lin et al., 2008; Yuan et al., 2009;
84 Zhu et al., 2011), even though they were merely short-term observations.

Many studies have also attempted to quantify transport fluxes of the main gaseous pollutants. A mobile laboratory study in Beijing city demonstrated regional transport of SO_2 from the NCP in both emission-control and non-control scenarios during the Beijing 2008 Olympics (Wang et al., 2011). Extrapolated from five 1-day case studies, the annual transport fluxes of SO_2 through the south-east part of the 6th Ring Road into Beijing were estimated at 49.2 Gg yr^{-1} and 146.3 Gg yr^{-1}, accounting for 70 % and 73 % of the annual SO_2 emissions in Beijing under emission-control and non-control scenarios, respectively. The Community Multi-scale Air Quality (CMAQ) model simulation by An et al. (2007) found that the regional transport from the surrounding areas of Beijing contributed 39 % of $PM_{2.5}$, 30 % of PM_{10}, and 18 % of SO_2 to the city on average in a heavy pollution episode in the spring of 2005. Similarly, the CMAQ model simulation over the Beijing region for July 2001, reported by Streets et al. (2007), illustrated the regional transport of $PM_{2.5}$ and O_3 between Beijing and the NCP. The study suggested that the average contributions of regional transport to $PM_{2.5}$ concentrations in the megacity Beijing from Hebei Province, Shandong Province, and Shanxi Province were about 32 %, 11 %, and 3.5 %, with maximum contributions of 70 %, 63 %, and 21 %, respectively. The regional transport contributions to the concentrations of O_3 in Beijing were less significant, with maximum contributions of 28 % from Hebei Province, 24 % from Shandong Province, and 10 % from Shanxi Province, respectively.

In summary, long-term observation of transport flux is necessary to constrain regional models and to directly evaluate the influence of regional transport on air quality. Estimations of air pollutant cross-boundary transport between Beijing and

the NCP are important for air quality management. However, evaluation of cross-boundary transport using long-term observations is very limited. In this study, we developed a method of calculating the surface transport flux intensity across a cross-boundary site based on long-term ground-based measurement and evaluated the regional transport influence of Beijing and the NCP on the cross-boundary site. The results showed different transport directions and seasonal variations in the surface transport flux intensities of the main pollutants, including SO_2, NO, NO_2, NO_x, O_3, and CO at the Yufa site. The key factors controlling regional transport are also discussed, which is important for the establishment of air quality control policy in future.

2. Measurements and Methods

2.1. Measurements

The Yufa site is located at the cross-boundary area between Beijing and the NCP and could be influenced by emissions from the megacity Beijing and long-range transport from the NCP. The measurements at the Yufa site (39°30'49"N, 116°18'15"E) were conducted on the top of a building (about 20 m above ground level) in the campus of Huangpu College. There is no tall building around the Yufa site which affects the wind and gaseous pollutant measurements. This is a rural site about 50 km south of the center of Beijing and near the border of Beijing Municipality and Hebei Province. As shown in Fig. 1, the Yufa site locates in the temperate monsoon climate zone and the topography of its surrounding area is flat. The prevailing wind of the Yufa site is the same as the surrounding region (Lin et al., 2009), thus the wind field of the Yufa site is representative of the researched area in this study. The northern and western sides

of the site are mountain areas where dry and clean air masses come from, whereas the southern and south-eastern sides are surrounded by heavily industrialised and urbanised areas, such as Hebei Province and Tianjin City (Fig.1).

Figure 1. here

The gaseous pollutant species measured included SO_2, NO, NO_2, NO_x, O_3, and CO. SO_2 was measured using a sulphur dioxide analyser (9850B; Ecotech, Knoxfield, Australia) which combines microprocessor control with pulsed UV fluorescence detection with the precision of 0.5 ppb and uncertainty within 10 %. The detection limit for the analyser is 0.5 ppb and the time resolution is 1 min. Reactive nitrogen species (NO, NO_2, and NO_x) were measured using nitrogen analyser (9841B; Ecotech) which utilises microprocessor control and chemiluminescence detection with the precision of 0.5 ppb and uncertainty within 10 %. The detection limit for the instrument is 0.5 ppb and the time resolution is 1 min. CO was measured using a CO analyser (9830A; Ecotech) which utilises NDIR Gas Filter Correlation photometry and microprocessor control with the precision of 0.1 ppm and uncertainty within 1 %. The detection limit for the instrument is 50 ppb and the time resolution is 1 min. O_3 was measured using an ozone analyser (9810B; Ecotech) which combines microprocessor control with UV photometry with the precision of 1 ppb and uncertainty within 5 %. The detection limit for the instrument is 0.4 ppb and the time resolution is 1 min. Measurements of meteorological parameters, including wind direction (WD), wind speed (WS), temperature (T), barometric pressure (BP), and relative humidity (RH), were conducted with a LASTEM auto meteorology station (LASTEM, Milan, Italy). All trace gas instruments were maintained and calibrated

routinely following the manufacturer's protocols. The main reasons for missing data were power and instrument failure. The detail information of the instruments was listed in Table 1.

Table 1. here

2.2. Methods

2.2.1. Transport direction analysis

The transport of gaseous pollutants is markedly influenced by meteorological parameters, especially wind speed and wind direction. For local emission sources, wind can facilitate the dilution and diffusion of air pollutants. Strong wind usually has marked diffusion capability, whereas weak wind usually leads to accumulation of air pollutants. For regional sources, strong wind can transport pollutants over long distances and may result in high concentrations of pollutants in downwind areas. Therefore, the relationship between pollutant concentration and wind field is an indicator of regional transport.

The bivariate polar plot graphical technique was used to investigate the relationships between the concentrations of gaseous pollutants and wind field, and to identify potential emissions sources and transport directions of air pollutants according to the technique developed by Carslaw et al. (2006) and Westmoreland et al. (2007). The variables (such as pollutant concentrations, wind speed, and wind direction) were plotted in polar coordinates. The procedure was as follows. First, the concentration data were partitioned into wind speed-wind direction bins, and the

mean concentrations were calculated within each bin. Then, the wind components *u* and *v* were calculated using Eq. (1):

$$u = \mathrm{WS} \cdot \sin(\pi\theta/180), v = \mathrm{WS} \cdot \cos(\pi\theta/180) \quad (1),$$

where *WS* is the hourly mean wind speed, and ϑ is the wind direction in degrees, with 90° being from the east. Then, a generalised additive model (GAM; Jayamurugan et al., 2013) was used for surface fitting to describe the concentration as a function of the wind components *u* and *v*. The concentrations calculated by the GAM can be expressed with Eq. (2):

$$\sqrt{C_i} = \beta_0 + s(u, v) + e_i \ (2),$$

where C_i is the calculated pollutant concentration, β_0 is the overall mean of the response, $s(u,v)$ is the smooth function, and e_i is the residual.

Compared to the nonparametric regression used by Henry et al. (2002), the bivariate polar plot involves the dependence of pollutant concentration on both wind speed and wind direction. The non-linear relationships among the variables (such as concentrations of gaseous pollutants, wind speed, and wind direction) as well as the interactions among these variables can be considered using the GAM method for data smoothing. In addition, the use of polar coordinates makes the graphics more intuitive.

2.2.2. Transport flux assessment

The surface transport fluxes at the Yufa site were calculated with the following formula (White et al., 1976; Wang et al., 2011):

$$f = -\frac{1}{n}\sum_{j=1}^{n} C_j \times WS_j \times cos\theta_j \quad (3),$$

196 $$\sigma = H_0 \times L_0 \quad (4),$$

197 $$FLUX = f \times \sigma \quad (5),$$

198 where f is surface flux intensity of the pollutants, i.e. the per unit area flux (μg s^{-1}
199 m^{-2}); C_j is the mean concentration of the pollutants (μg m^{-3}) during the jth
200 observation hour; ϑ_j is the angle between wind direction and the north-south
201 direction during the jth observation hour; and WS_j is wind speed (m s^{-1}) during the
202 jth observation hour; n is the total number of observation hour; σ is the surface
203 cross-sectional area (m^2) with the width of L_0 (m) and height of H_0 (m); the average
204 surface flux of the pollutants (i.e. *FLUX*, μg s^{-1}) can be obtained by multiplying flux
205 intensity f and the cross-section area σ.

206 Figure S1 shows a schematic diagram of the surface flux calculation. The flux
207 intensity here is the product of wind vector and air pollutant concentration
208 measured at the same location. Ideally, we need to use the wind speed and air
209 pollutant concentration with infinite small time resolution to conduct the surface flux
210 calculations. In this study, the hourly data of the pollutants and wind were used,
211 mainly because the pollutants concentration data was converted from the minutes'
212 data to hourly mean to remove the accidental fluctuation and reduce the noise.
213 Therefore, we assumed the wind speed and wind direction were constant within one
214 hour, and hourly wind data was used to match with the hourly air pollutant
215 concentration data to calculate the flux intensity.

216 It also need to make it clear that the surface flux intensity calculated in this study
217 is the per unit area flux across the Yufa site, which is different from the flux across a
218 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, wind speed and direction were homogenously distributed, vertically and horizontally. Otherwise, vertical profiles of air pollutants concentration and wind are needed to calculate the cross-section transport flux of two adjacent regions for the whole boundary layer with the integrating formula below:

$$FLUX = \iint C_{(x,z)} WS_{(x,z)} sin\theta_{(x,z)} dxdz = \iint f_{(x,z)} dxdz \quad (6),$$

where x is horizontal distance to the observed point, z is the vertical distance from ground to the observed point. In this study, we focus on the method developing of the surface flux intensity calculation and evaluation of the regional transport influence of Beijing and the NCP on the cross-boundary site based on the ground-based observation data.

2.2.3 The backward trajectory model and PSCF analysis

The 12 h air mass back trajectories arriving at the Yufa site at 500 m above the ground level were calculated using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory Version 4 model (HYSPLIT-4 model) (http://ready.arl.noaa.gov/HYSPLIT.php) during the study period (from 15 August 2006 to 31 October 2008) with a 1 ˚ × 1 ˚ latitude-longitude horizontal resolution and the final meteorological database. The final archived meteorological data was obtained from the National Center for Environmental Prediction's (NCEP's) Global Data Assimilation System (GDAS) (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1). The back trajectories were

generated with 6-h time resolution (four times per day) at starting times of 0:00, 6:00, 12:00 and 18:00 UTC (8:00, 14:00, 20:00, 4:00 LT-local time, respectively).

The potential source contribution function (PSCF) analysis was performed with the Gis-based software TrajStat (http://www.meteothinker.com/products/trajstat.html) (Wang and Zhang et al., 2009). The PSCF analysis has been widely used for identifying the possible source areas of the observed high concentrations of pollutants at the receptor site (Ashbaugh et al., 1985; Zhang et al., 2013). In this study the long-term calculated surface flux intensity data was assigned to the backward trajectories in the PSCF analysis to confirm the bi-directional transport of pollutants between Beijing and the NCP. The PSCF analysis was conducted as follows.

The study domain was divided into i × j equal size grid cells and the PSCF value for *ij*th cell is defined as:

$$\mathrm{PSCF}_{ij} = m_{ij} / n_{ij} \quad (7),$$

where n_{ij} denoted the number of endpoints that fall in the *ij*th cell, and m_{ij} represented the number of endpoints for the same cell having arrival times at the observed site corresponding to measured data higher than an arbitrarily set criterion.

To reduce the effect of small values of n_{ij}, the PSCF values were multiplied by an arbitrary weigh function W_{ij}. In this study, W_{ij} is defined as below.

$$W_{ij} = \begin{cases} 1.00, & 80 < n_{ij} \\ 0.70, & 20 < n_{ij} \leq 80 \\ 0.42, & 10 < n_{ij} \leq 20 \\ 0.05, & n_{ij} \leq 10 \end{cases} \quad (8),$$

In this study, the study domain was 30–50 ˚ N, 100–125 ˚ E and the horizontal resolution was 0.25 ˚ × 0.25 ˚.

3. Results and discussion

3.1. Observations

Figure 2. here

The time series of hourly average and 24-hour smoothing concentrations of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO observed at the Yufa site from 15 August 2006 to 31 October 2008 are shown in Fig. 2. The hourly mean ± SD (median) concentration value of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO was 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 indicated that the Yufa site has become a relatively polluted rural site. Typical seasonal variations were observed for all gaseous pollutants. Concentrations of primary pollutants, including SO_2, NO, NO_2, NO_x, and CO, were high in winter and low in summer. In contrast, the concentration of O_3, which is a secondary pollutant, was high in summer and low in winter.

Figure 3. here

Meteorological parameters such as WS, WD, RH, T, and BP were also measured at the Yufa site; the monthly statistics are shown in Fig. 3. North (usually in winter) or south wind (usually in summer) prevailed at the Yufa site, with monthly average wind speed mostly below 2 m s^{-1}. Exceptional conditions occurred occasionally in spring and winter for the north wind, with monthly average wind speeds around 2–3 m s^{-1}. In addition, for the north wind, the mean speed was higher than the median speed, suggesting the prevalence of high wind speeds in both spring and winter. Prevailing north wind with high wind speed during winter and spring has been reported consistently in the Beijing area (Lin et al., 2008; Wehner et al., 2008). Another exceptional condition occurred in spring for the south wind, with a monthly average wind speed around 2 m s^{-1}. Figure 4 summarises the prevalence of wind direction in the four seasons. Generally, the prevailing surface wind directions were north-northeast and south-southwest in all seasons. In winter and spring, winds from the north-northeast sector made a contribution of about 40–50 % to wind frequency. Whereas under the influence of summer monsoon, winds from south increased significantly in summer, with the contribution to wind frequency above 40 %. RH was higher in summer and lower in spring and winter with the driest month in April of 2007 and February of 2008. The seasonal variation of RH may partially be related to the variations of WS (Lin et al., 2011). T was higher in summer and lower in winter. Surface pressure measurements showed high values in winter and low values in summer due to surface heating and lifting air masses in summer, which partly accounted for the wind field in the NCP (Takegawa et al., 2009).

Figure 4. here

305 The seasonal variations in gaseous pollutants and meteorological parameters
306 could be linked in certain ways. For example, the high temperature and low pressure
307 in summer suggested a high boundary layer and diluted gaseous pollutants to some
308 extent. The high temperature, light intensity, and relative humidity also favoured the
309 chemical transformation of these primary pollutants and the formation of secondary
310 pollutants. The high wind speeds in spring and winter also affected regional
311 transport, and therefore the concentrations of gaseous pollutants, as discussed
312 below.

313 **3.2. Transport direction**

314 **3.2.1. The bivariate polar plots for the whole observed period**

315 As shown in Fig. 1, the Yufa site is located at the boundary area of Beijing city and the
316 NCP. Prevalent south/south-west or north/north-east wind would bring in polluted or
317 clean air masses to the site. Air masses from both directions would pass over the
318 Yufa site. Regional transport from the megacity Beijing and the NCP could therefore
319 be observed at the Yufa site. The transport directions for gaseous pollutants,
320 including SO_2, NO, NO_2, NO_x, O_3, O_x, and CO, will be discussed in this section.

321 Figure 5. here

322 Figure 5a-g show the bivariate polar plots for SO_2, NO, NO_2, NO_x, O_3, O_x, and CO
323 at the Yufa site, respectively. In the low wind speed scenario, high or medium
324 concentrations of NO, NO_2, NO_x, SO_2, and CO were generally observed, along with
325 low O_3 and O_x concentrations. In the high wind speed scenario, the dependence of
326 species concentration on wind speed and wind direction was more varied.

Specifically, the bivariate polar plot in Fig. 5b clearly shows dependence of high NO concentration (higher than 30 ppb) on low wind speed, with low NO concentration (lower than 5 ppb) at wind speeds > 3 m s^{-1}. The bivariate polar plot in Fig. 5c shows similar dependence of high NO_2 concentration on low wind speed, but NO_2 concentrations up to 20 ppb were still observed with medium wind speeds of around 5 m s^{-1} from the south, east, and north-east. Accordingly, the dependence pattern of the NO_x concentration (Fig. 5d) on wind speed and wind direction reflected the features of both NO and NO_2. The dependence pattern of high CO concentration on low wind speed in Fig. 5g was similar to that for NO_x, but a considerable CO concentration, substantially higher than background level, was still observed at wind speeds exceeding 5 m s^{-1} from the south and the east. Figure 5a shows similar dependence of medium-high concentration of SO_2 (around 20 ppb) on low wind speed, with one unique feature being that high SO_2 concentration was observed under conditions of high wind speed (> 5 m s^{-1}) in various wind directions (especially the south wind). Finally, the bivariate polar plot in Fig. 5e shows the dependence of O_3 concentration on wind speed and wind direction, which was somewhat opposite to the patterns for other species. The low O_3 concentration (< 20 ppb) was related to low wind speed or calm wind conditions. With the north wind and medium or high wind speed, a typical background O_3 concentration (around 50 ppb) was observed. With south wind and medium or high wind speed, high O_3 concentration was observed. The dependence of the high O_x concentration on high wind speed from the south and south-east was similar to that of O_3, but no low concentration of O_x was

observed under low wind speed conditions (Fig. 5f), probably due to the compensation of high NO_x concentration at low wind speeds (Fig. 5d).

Figure 6. here

The high concentrations of NO, NO_2, NO_x, and CO and the medium-high concentration of SO_2 observed under low wind speed conditions were consistent with their high emission intensities in the Beijing area (Fig. 6). Due to the marked increase in the number of vehicles and heavy energy consumption, Beijing has been a well-known emission hot spot for NO and NO_2 (Tang, 2004). Meanwhile, the extremely high levels of CO emissions in the Beijing area are clearly shown in the emissions map (Fig. 6) and have been reported consistently (Wang et al., 2009) and directly observed, with peak CO concentrations up to 9.3 ppm. Only medium-high SO_2 concentration (~15 ppb) observed even at low wind speed suggested the successful reduction of SO_2 emission, which could be ascribed to the continuous effort of the Chinese government since the 1990s and during the Olympic Games (Qin et al., 2009; Tang, 2004; Wang et al., 2009, 2011). Accordingly, the O_3 concentration under low wind speed conditions was lower than the typical background level, which could be attributed to the rapid titration of O_3 by of accumulation NO.

3.2.2. Seasonal variations of the bivariate polar plots

Figure 7. here

The different patterns of the bivariate polar plots reflected the differences in local emission and regional transport for different species. The emissions, the

meteorological conditions, the chemical reaction rate and the species lifetime, which have essential influence on the regional transport, vary greatly by seasons. Thus the seasonal variations of the bivariate polar plots and the corresponding causes were discussed in this section.

Figure 7b-d show seasonal variations of the bivariate polar plots for NO, NO_2, and NO_x at the Yufa site, respectively. Generally, the mean concentrations of NO_2, NO_x and especially NO in the low wind speed scenario were higher than those in the higher wind speed scenario in all seasons. The mean concentration of NO was less than 10 ppb when the wind speed higher than 5 m s^{-1} in all seasons (Fig. 7b). Figure 7c clearly shows the relatively higher concentration of NO_2 (~20 ppb) with winds at higher wind speed (> 5 m s^{-1}) from the south sector in spring, from the northeast and south sectors in summer and winter, and from the northeast sector in autumn. Figure 7d shows the dependence pattern of NO_x was similar to both NO and NO_2. Although emission hot spots of NO, NO_2, and NO_x are widespread in the NCP, the long-range transport of these species to Yufa is limited by the lifetime of these species. As the average O_3 concentration for spring, summer, autumn and winter was 20, 11, 32, and 42 ppb respectively at Yufa, the typical lifetime of NO was 66, 51, 106, and 181 s in spring, summer, autumn, and winter, respectively, just by assuming that all the NO is removed mainly by chemical reaction with O_3 (Sander et al., 2011). The transport distance of NO was therefore less than 5 km even with a high wind speed of 15 m s^{-1}. Even when considering the 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). That is, NO concentration is determined by local emissions rather than regional transport. NO_2 and NO_x have longer lifetimes in the atmosphere than NO has, typically on the order of 4–5 h and with longer photochemical lifetime in cold seasons (Beirle et al., 2011; Gu et al., 2013). Hence, the typical transport distance of these species is around 100 km at the wind speed of 5 m s^{-1} (Beirle et al., 2011). Within such transport distance, the Yufa site is surrounded by various NO_x emission hot spots (Fig. 6), such as the megacity Beijing to the north, the Baoding-Cangzhou area to the south, and the Tianjin-Tangshan area to the east. Meanwhile the emission intensity was larger in winter and autumn than that in spring and summer (Fig. 6). It is therefore reasonable to observe the influence of short-range transport, in addition to local emissions, on the local NO_2 and NO_x concentrations, especially in cold seasons (Fig. 7c and 7d). Although our results suggested that short-range transport from these surrounding areas, especially the urban area of Beijing, was a non-negligible factor affecting the NO_x concentration at the Yufa site, the regional transport of NO_x was of less significance compared to SO_2 and CO due to its limited transport distance (see below).

Figure 7e is the seasonal bivariate polar plots of CO, which clearly shows the relatively higher mean concentration of CO (> 1 ppm) with winds at low wind speed (< 2 m s^{-1}), similar to nitrogen oxide species. The mean concentration of CO was relatively higher with wind at higher wind speed (> 5 m s^{-1}) from south sector in spring and summer, from northeast and south sector in autumn, and from north and south in winter. The oxidation lifetime of CO is typically ~20 days, under the assumption of OH radical concentration of 2×10^6 cm^{-3} (Xu et al., 2011). This is

417 substantially longer than the lifetime of NO_x, making regional transport of CO an
418 important process affecting local air quality in the downwind area. The different
419 lifetimes of CO and NO_x appeared to explain the unique high concentration of CO,
420 but not NO_x, at wind speeds exceeding 5 m s^{-1} from the south and the east. Our
421 results suggest that regional transport from the south and central NCP and the
422 Tianjin area could greatly affect local concentrations of CO at the Yufa site.

423 Figure 7a clearly shows the relatively higher mean concentration of SO_2 (~20 ppb)
424 with winds at higher wind speed (> 5 m s^{-1}) from the south sector in spring and
425 summer. The mean concentration of SO_2 was high (> 30 ppb) with winds at higher
426 wind speed (> 5 m s^{-1}) from the north-east, east, and south sectors in autumn and
427 winter. Similar to CO, SO_2 has a relatively long lifetime in the atmosphere compared
428 to NO_x, i.e. a couple of hours to 1–2 days with longer lifetime in winter and shorter
429 lifetime in summer (Beirle et al., 2014; He et al., 2012; Lee et.al., 2014), and regional
430 transport of SO_2 was expected to occur. Accordingly, regional transport from
431 emission hot spots located south of the Yufa site (Fig. 6), was found to influence the
432 concentrations of SO_2 (Fig. 7a) at Yufa in all seasons. Specifically, the highlighted
433 emission hot spots in the central NCP and the south NCP, which accounted for about
434 70 % of China's coal consumption in 10 % of China's domestic area (China Statistical
435 Yearbook, 2008), is a major source of SO_2 in the Beijing area by regional transport
436 (Liu et al., 2016). Furthermore, regional transport from the north-east sector of the
437 Yufa site, where the center of the megacity Beijing located, also was observed in
438 autumn and winter, which indicated the increased emission of SO_2 in heating
439 seasons.

Finally, the bivariate polar plots in Fig. 7f and 7g show the dependence of O_3 and O_x concentration on wind speed and wind direction by season. The low O_3 concentration (< 20 ppb) was observed at low wind speed (< 2 m s^{-1}). With the north wind at higher wind speed (> 5 m s^{-1}), a typical background O_3 concentration (around 50 ppb) was observed in spring and summer. With south wind at higher wind speed (> 5 m s^{-1}), high O_3 concentration (above 60 ppb) was observed, especially in summer. The main difference of seasonal bivariate polar plots between O_3 and O_x was that no low concentration of O_x was observed under low wind speed conditions in all seasons. The low concentration of O_3 at low wind speed may be due to the titration of O_3 by NO, which was more obvious in autumn and winter. Background O_3 levels in the north-west wind under medium and high wind speed conditions clearly reflect the transport of background air mass to the Yufa site from locations where the emission intensities of pollutants were relatively low (Fig. 6), and this was more obvious in spring when the air masses from the north-west increased (Fig. 4). Whereas O_3 concentrations higher than background level in the south wind under medium and high speed conditions, especially in summer, suggest accumulation of O_3 during its transport from the central NCP area or even the south NCP area to the Yufa site. Emission intensity of O_3 precursors, such as NO_x and VOCs is high in the NCP, and the solar radiation is strong in summer, which facilitate the formation and transport of O_3 from the NCP to Beijing (Zhang et al., 2014).

In conclusion, the emissions in the Beijing area are closely related to the observed concentrations of NO, NO_2, NO_x, and CO at Yufa. Regional transport had a clear influence on the concentrations of all gaseous pollutants examined here, with

463 the exception of NO. The emission hot spots located east, north-east, and especially
464 south of the Yufa site determined the regional transport directions. The influence of
465 regional transport differed among species. Regional transport of SO_2, CO, and O_3
466 from the central and south NCP to the Yufa site was more important, whereas
467 regional transport of NO_x from the NCP was less evident. Factors affecting regional
468 transport included, but were not limited to, the atmospheric lifetime of pollutants,
469 wind field, and local and regional emissions. As the Yufa site is a cross-boundary rural
470 site between the megacity Beijing and the NCP, observation of transport flux there is
471 appropriate in evaluating the regional transport influence by both the megacity
472 Beijing and the NCP on the Yufa site.

473 **3.3. Transport flux**

474 To evaluate the surface transport of the main air pollutants from Beijing and the
475 NCP to the Yufa site, the surface flux intensities were calculated with Eqs. (3) based
476 on observations at the Yufa site. The mean net surface flux intensities in each
477 season were also calculated for the 2-year observation period (Table 2). The overall
478 net surface flux intensities (mean ± SD) of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO were
479 6.2 ± 89.5, -4.3 ± 29.5, -0.6 ± 72.3, -4.9 ± 93.0, 14.7 ± 187.8, 14.8 ± 234.9, and 70 ±
480 2830 μg s^{-1} m^{-2} during the observation period from 01 September 2006 to 31
481 August 2008, respectively. The large standard deviation of the surface flux intensity
482 indicated the large variations of the transport flux intensities. Table 3a shows the
483 mean influx intensities (positive; from the NCP to Yufa) were highest in winter and
484 lowest in summer, with the flux intensity values in winter 2–6 times of those in
485 summer. The outflux intensities (negative; from Beijing to Yufa) show the same

pattern, with the absolute flux intensity values in winter 2–8 times of those in summer (Table 3b). Yet the overall net transport surface flux intensities show quite different seasonal variations (Table 2) comparing to the results in Table 3. For SO_2, CO, O_3, and O_x the surface transport flux intensities from the NCP to Yufa surpassed those from Beijing to Yufa in all seasons except in winter, with the strongest net fluxes largely appeared in summer, which was about 4–8 times of other seasons. The net surface transport flux intensity of NO_x from Beijing to Yufa was stronger than that from the NCP to Yufa except in summer, with the strongest net flux in winter, which was about 1.3–8 times of other seasons.

Table 2. here

Table 3. here

To understand the transport fluxes reported here, it is necessary to discuss the affecting factors. First, the prevalent wind is a dominant factor affecting the surface fluxes. Figure 8 shows the time series of daily average surface flux intensity, i.e. the per unit cell flux (μg s^{-1} m^{-2}) of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO, and corresponding wind vectors (m s^{-1}) during the observation period. In general, the variations in the pollutant flux intensities showed a saw-teethed pattern, with influx (positive; from the NCP to Yufa) and outflux (negative; from Beijing to Yufa) prevailing according to the shift in wind direction. Meanwhile, mainly due to the seasonal variations in wind speed and wind direction (Fig. 3 and 4), the magnitude of surface fluxes showed similar seasonal variation (Table 2). High net positive influx intensities were observed in summer, and high net negative outflux intensities in

508 winter. As the north wind prevailed significantly over the south wind in winter, and
509 the south wind over the north wind in summer (Fig. 4), the values of net surface
510 flux intensities in these two seasons were the highest. During the other two
511 seasons, frequent changes in positive and negative fluxes tended to cancel each
512 other out, making the net transport fluxes less significant. This dominant role of
513 wind field could also be illustrated by conditions during the winter of 2006/07 and
514 2007/08. Exceptionally, the south wind prevailed in the winter of 2006/07 (Fig. 4),
515 leading to the surface flux intensity of pollutants more positive in the winter of
516 2006/07 than 2007/08 (Table 2). For example, the increase of influx intensity for
517 SO_2, NO, NO_2, NO_x and CO between winter of 2006/07 and 2007/08 was on the
518 order of a factor of 1.5 (Table 3a).

519 Figure 8. here

520 Second, the transport flux is determined not only by the wind field but also by
521 the emissions of pollutants in the upwind area. Various pollutants showed different
522 patterns of seasonal variations in flux as a result of relative high emission intensities
523 in the upwind area compared to local emissions. For example, the seasonal surface
524 flux intensities of SO_2 were mainly positive influx, except in winter of 2007/08. The
525 significant regional transport of SO_2 from the NCP to Yufa in all seasons except
526 winter could be partly attributed to the high emission intensity of SO_2 in the NCP
527 (Fig.6) and the reduction of SO_2 emission in Beijing (Qin et al., 2009; Wang et al.,
528 2009, 2011), whereas the SO_2 outflux from Beijing to Yufa was determined by the
529 prevalent north wind, as explained above. In contrast to the net positive influx of
530 SO_2, the net seasonal surface flux intensities of CO were negative in both winter

and autumn. The small outflux of CO in autumn reflected increased CO emission in Beijing, which was sufficiently strong to account for the strong CO emissions in the NCP.

The influence of emissions on transport flux could also be inferred from an emissions-reduction scenario. For example, the 29th Olympic Games was held in Beijing during the period from 8 August 2008 to 20 September 2008. The Beijing government implemented aggressive long- and short-term air quality control measures in Beijing and its surrounding areas before and during the Olympic period to maintain good air quality during the Olympic Games (Wang et al., 2010; Wang et al., 2011). The control measures included moving the heavy polluted factories out the Beijing city, reducing the traffic emission through an odd/even plate number rule, and freezing construction activities (Wang et al., 2009). The concentrations of pollutants and the surface flux intensities during the 2008 Olympic Games were substantially reduced compared to the corresponding period of 2007 (Table 4). Besides the favored meteorological conditions (Fig. S2), the significant emission reduction both in the Beijing area and the NCP during the 2008 Beijing Olympic Games played a key role in the decrease of the transport flux intensities (Zhou et al., 2010).

Table 4. here

Finally, the chemical properties of these species could also affect the flux. Take O_3 for example, although both Beijing and the NCP are regarded as emissions hot spots for O_3 precursors, the short distance between Beijing and the Yufa site may hinder the secondary formation of O_3 to some extent. Thus the surface transport of

554 O_3 from to the NCP to Yufa was stronger than that from Beijing to Yufa, especially in
555 summer time with a net average surface flux intensity value of about 60 μg s^{-1} m^{-2},
556 which is 4–9 times of that in autumn and spring (Table 2). The lifetime of the
557 pollutants also determined the different net transport flux intensities for different
558 species (Table 2 and 3), with the net transport of NO, NO_2 and NO_x from Beijing to
559 Yufa and the net transport of SO_2, CO, O_3 and O_x from the NCP to Yufa. These results
560 are consistent with bivariate polar plots analysis mentioned above (Fig. 5 and 7).

561 Overall, the flux intensities are influenced by at least the wind field, emissions
562 inventory in both the megacity Beijing and the NCP, and the chemical fates of these
563 pollutants in the atmosphere. These observations provide insight for the analysis of
564 projected transport flux under various emissions-reduction scenarios in the future.
565 On the other hand, the dependence of the fluxes on these factors, which can vary,
566 suggests that the fluxes reported here should not be compared with other reports
567 under different conditions.

568 **3.4 The back trajectory and PSCF analysis**

569 The discussion above suggested the regional transport from both Beijing and the
570 NCP have important influence on the air quality of the Yufa site. However, both the
571 bivariate polar plots and surface flux intensity calculation were based on the
572 observation data at a ground measurement site. Considering the limitation of
573 spatial representation of the Yufa site, the PSCF analysis based on the HYSPLIT-4
574 model was used to demonstrate the regional transport influence of the megacity
575 Beijing and the NCP on Yufa in this section.

Figure 9. here

Figure 10. here

PSCF analysis was used in this study by combining backward trajectories and the corresponding surface transport flux intensities of pollutants. PSCF results of SO_2, NO, NO_2, NO_x, CO, O_3 and O_x in 6-h time resolution are shown in Fig. 9 for positive influx intensities (i.e. from south to north) and Fig. 10 for negative outflux intensities (i.e. from north to south). It can be seen from Fig. 9, that the higher PSCF values for most pollutants are located at the area south-west to the Yufa site, which indicates the positive surface flux intensities of the Yufa site are consistent with the air masses moving from south to Yufa. Figure 10 shows the higher PSCF values for most pollutants are located at the area north to the Yufa site, which indicates the negative surface flux intensities of the Yufa site are consistent with the air masses moving from north to Yufa. The PSCF analysis results validate the calculated flux intensities based on observation data can be used to evaluate the regional transport influence of Beijing and the NCP on the Yufa site. However, it should be noticed that the PSCF results of NO, NO_2, and NO_x was inconsistent with the flux calculation results sometimes (Fig. 9 and 10), which may partially ascribe to the lifetime of these species is much shorter than 12 h. As a cross-boundary site between the megacity Beijing and the NCP, the surface flux intensities at the Yufa site may also indicate the transport between the megacity Beijing and the NCP.

3.5 Uncertainty and limitation

Uncertainty in calculation of the surface flux intensities in this study mainly comes from the measurement of the pollutants and the wind. Based on the instruments used, the uncertainty of the measurement of the concentrations of SO_2, NO_x, CO, and O_3 was within 10 %, 10 %, 1 %, and 5 %, respectively. The uncertainty of wind speed measurement was less than 5 % and the uncertainty of wind direction was about 1 %. Thus, the uncertainty of the overall surface flux intensity for SO_2, NO_x, CO, and O_3 was less than 12 %, 12 %, 6 %, and 8 %, respectively.

In this study, we did not intent to extrapolate from the Yufa site to the entire region. We 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. Bivariate Polar plots analysis and surface flux intensity calculation were conducted, and we obtained clear evidences 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 the influence of the boundary layer on the regional transport, three dimensional observations with high precision and resolution are needed for further comprehensive discussion of the regional transport between Beijing and the NCP.

4. Conclusions

We used 2-year continuous observation data at a cross-boundary rural site between the megacity Beijing and the NCP to investigate regional transport influence on the Yufa site, as part of the "Campaign of Air Quality Research in Beijing and Surrounding Region 2006–2008" (CAREBeijing 2006–2008). The gaseous pollutants SO_2, NO, NO_2,

620 NO_x, CO, O_3, and O_x, together with meteorological data, were determined at Yufa
621 from August 2006 to October 2008. During the observation period, the average
622 concentrations of the pollutants at the Yufa site were relatively high, suggesting a
623 profound influence of the emissions from the megacity Beijing and regional transport
624 from the NCP.

625 Through bivariate polar plots, we found that the south wind, at relatively high
626 wind speed, was essential for the inflow of SO_2, CO, and O_3 from the NCP to Yufa. For
627 NO, NO_2, NO_x, and even CO, the emission from Beijing played a dominant role. The
628 seasonal variations of emission intensity, meteorological conditions, pollutant
629 lifetimes lead to the seasonal variations of the regional transport of pollutants, hence
630 the different bivariate polar plot patterns.

631 The the surface flux intensities showed strong net surface transport from the
632 NCP to Yufa in summer and net surface transport from Beijing to Yufa in winter,
633 mainly varied with the prevailing wind. The positive net influxes of SO_2, CO, and O_3
634 in this study indicate a northward regional transport of these species from the NCP.
635 Whereas the fluxes of NO_x indicate the influence of NO_x emission in Beijing city
636 could only influence downwind area adjacent to Beijing, due to the limited
637 transport distance of NO_x.

638 PSCF analysis demonstrated the regional transport from Beijing and the NCP to
639 Yufa can be evaluated by the surface flux intensity calculation based on the ground-
640 based measurement data. As a cross-boundary site between the megacity Beijing
641 and the NCP, the surface transport flux intensities at the Yufa site may also indicate
642 the transport between the megacity Beijing and the NCP.

643 Our results again suggested that Beijing and the NCP have tight interactions
644 through regional transport of air pollutants. Factors affecting the transport flux such
645 as meteorological parameters, especially wind speed and wind direction, emissions
646 inventory, and photochemical reactions are essential for the regional transport fluxes
647 and thus the air quality of the megacity Beijing and its surrounding areas. Therefore,
648 both local emissions reduction and regional cooperative control should be taken
649 considered in air quality management of Beijing.
650
651 **Author contribution**. T. Zhu designed the experiments and L. Zeng and the staff of
652 the Yufa site carried out the experiment. Y. Li conducted the data analysis with
653 contributions from all co-authors. J. Liu provided the emission maps. J. Wang
654 managed the observation data of the program. Y. Li prepared the manuscript with
655 the help of T. Zhu, C. Ye, J. Liu and Y. Zhu.
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828 **Table 1.** The overview of measurement instruments.

Species/ Parameter	Instrument	Detection limit	Time resolution	Precision	Uncertainty
SO_2	Ecotech 9850B	0.5 ppb	1 min	0.5% (0.5 ppb)	10%
NO-NO_x	Ecotech 9841B	0.5 ppb	1 min	1% (0.5 ppb)	10%
CO	Ecotech 9830	50 ppb	1 min	1% (0.1 ppm)	1%
O_3	Ecotech 9810B	0.4 ppb	1 min	0.5% (1 ppb)	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 °C	±0.2°C
RH	LASTEM	-	10 min	1%	±3%

829

830 **Table 2.** The total and seasonal net surface flux intensities (mean ± SD) (μg s^{-1} m^{-2}) of

831 gaseous pollutants at the Yufa site from 1 September 2006 to 31 August 2008.

Flux (μg s^{-1} m^{-2})	SO_2	NO	NO_2	NO_x	CO	O_3	O_x
Autumn_06	5.3±79.6	-6.3±27.5	-3±60.2	-9.4±78.9	-30±2730	19.4±128.8	25.9±177.6
Autumn_07	6.3±78.8	-6.6±33.6	-3.5±74.3	-10.1±98.6	-60±2570	10±120	6.6±170.3
Winter_06/07	11.8±139.1	-6.9±47.7	3.6±105.6	-3.3±142.9	350±4150	-11.9±127.5	-8.3±188
winter_07/08	-13.1±113.3	-11.5±46.5	-11±82.3	-22.6±117.6	-550±3380	-29.6±143.1	-40.7±191.2
Spring_07	11.3±90.5	-1.9±12.1	0.1±71.1	-1.9±78.8	50±2720	3.4±261.8	3.5±315.5
Spring_08	13.5±92	-1.8±15.2	0.2±76.4	-1.5±87	160±2630	10.7±266.4	10.9±321.2
Summer_07	11±35.3	0.4±6.7	8.4±46.9	8.7±51.5	600±1960	71.3±175.7	79.7±211.4
Summer_08	5.7±26.1	0.1±4.9	1±32.7	1.1±36.6	120±1540	48.1±183.3	49.1±207.8
Total	6.2±89.5	-4.3±29.5	-0.6±72.3	-4.9±93	70±2830	14.7±187.8	14.8±234.9

832

833 **Table 3a.** The total and seasonal surface influx intensities (mean±SD) (positive; from

834 the NCP to Yufa, μg s^{-1} m^{-2}) of gaseous pollutants at the Yufa site from 1 September

835 2006 to 31 August 2008.

Influx (μg s^{-1} m^{-2})	SO_2	NO	NO_2	NO_x	CO	O_3	O_x
Autumn_06	53.3±62.7	5.1±7.9	42.5±34.3	47.6±37.9	1770±1740	85±131.6	143.1±158.3
Autumn_07	43.9±80.8	8.5±14.5	48.7±51.8	57.2±60.7	1720±1820	65.1±116.8	117.3±140.2
Winter_06/07	106.3±126.1	19.8±30	82.3±83	102±101.5	3360±3620	40.1±74.1	122.4±112.8
winter_07/08	72.3±95.1	13.6±20.9	60.4±60.2	74±72.6	2170±2130	41.7±70.2	102.1±98.3
Spring_07	62.9±88.3	3.8±7.2	53.6±46.6	57.5±49.7	1970±2050	158.8±193.2	212.5±223.1
Spring_08	64.7±97.5	6±9	56.8±44.6	62.7±49.1	2090±1850	162.5±194.4	222.9±217.9
Summer_07	22.3±38.7	2.9±5.4	32.4±30.1	35.3±32.4	1560±1630	140.7±177.9	173.1±199.7
Summer_08	18.9±24.3	2.8±2.6	20.8±16.3	23.6±18.2	1060±880	138.2±168.9	160.1±180.2
Total	53.2±84.8	7.4±15.4	48.3±51.5	55.8±60.7	1920±2130	108.2±159.2	159.7±180

836 **Table 3b.** The total and seasonal outflux intensities (mean ± SD) (negative; from

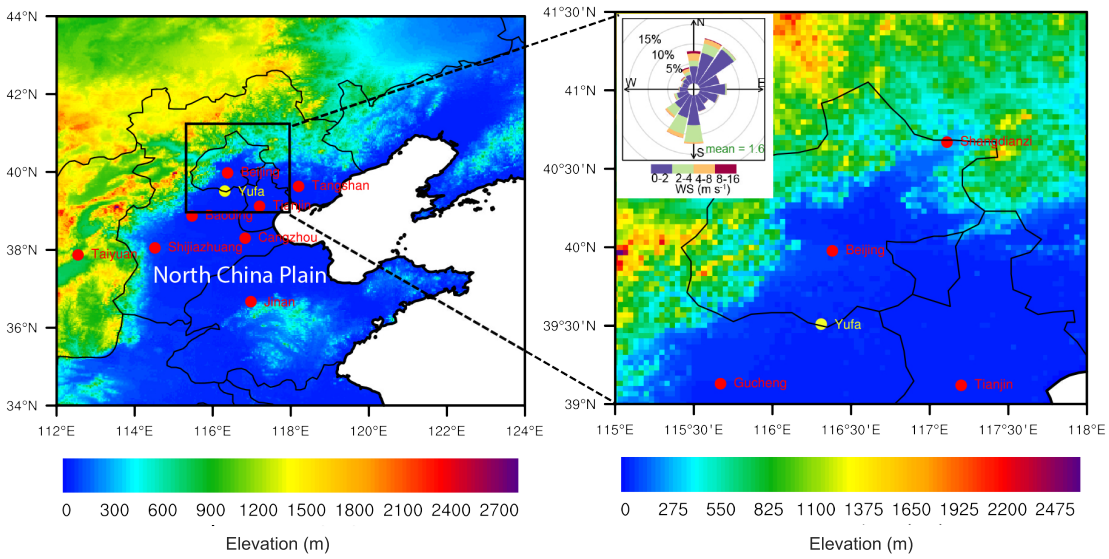
837 Beijing to Yufa, μg s^{-1} m^{-2}) of gaseous pollutants at the Yufa site from 1 September

838 2006 to 31 August 2008.

Outflux ($\mu g\ s^{-1}\ m^{-2}$)	SO_2	NO	NO_2	NO_x	CO	O_3	O_x
Autumn_06	-40.4±66	-17.8±34.4	-48.6±43.9	-66.4±67.3	-1830±2360	-44.7±87.5	-91.2±103.4
Autumn_07	-30.2±56.5	-21.4±39.9	-54.5±54.9	-75.9±82.9	-1800±1910	-48.9±92.3	-101.5±120.4
Winter_06/07	-72.8±86	-30.7±48.1	-66.8±67.6	-97.6±103.8	-2350±2380	-58.4±146.3	-125.2±163.9
winter_07/08	-73.9±81.6	-29.5±51.2	-61.9±52.9	-91.3±92.4	-2490±2690	-80.4±159.2	-142.3±175.7
Spring_07	-41.3±55.8	-7.8±13.2	-54.6±45.6	-62.4±52.6	-1920±1720	-155.2±225	-209.8±245.6
Spring_08	-38.8±44.4	-9.7±16.2	-57.4±56.4	-67.1±65.8	-1820±1660	-151.3±235.8	-205.2±259.7
Summer_07	-9±13.3	-4.2±6.5	-34.2±40.8	-38.4±44.9	-1110±1210	-51.6±76.6	-85.8±102.2
Summer_08	-12.1±15.8	-3.5±5	-25.6±30.1	-29.1±33.3	-1150±1320	-75.2±119.5	-100.1±137.1
Total	-42.8±64.6	-16.7±35.2	-52±52.8	-68.7±77.1	-1870±2080	-85±163.2	-137.3±184.5

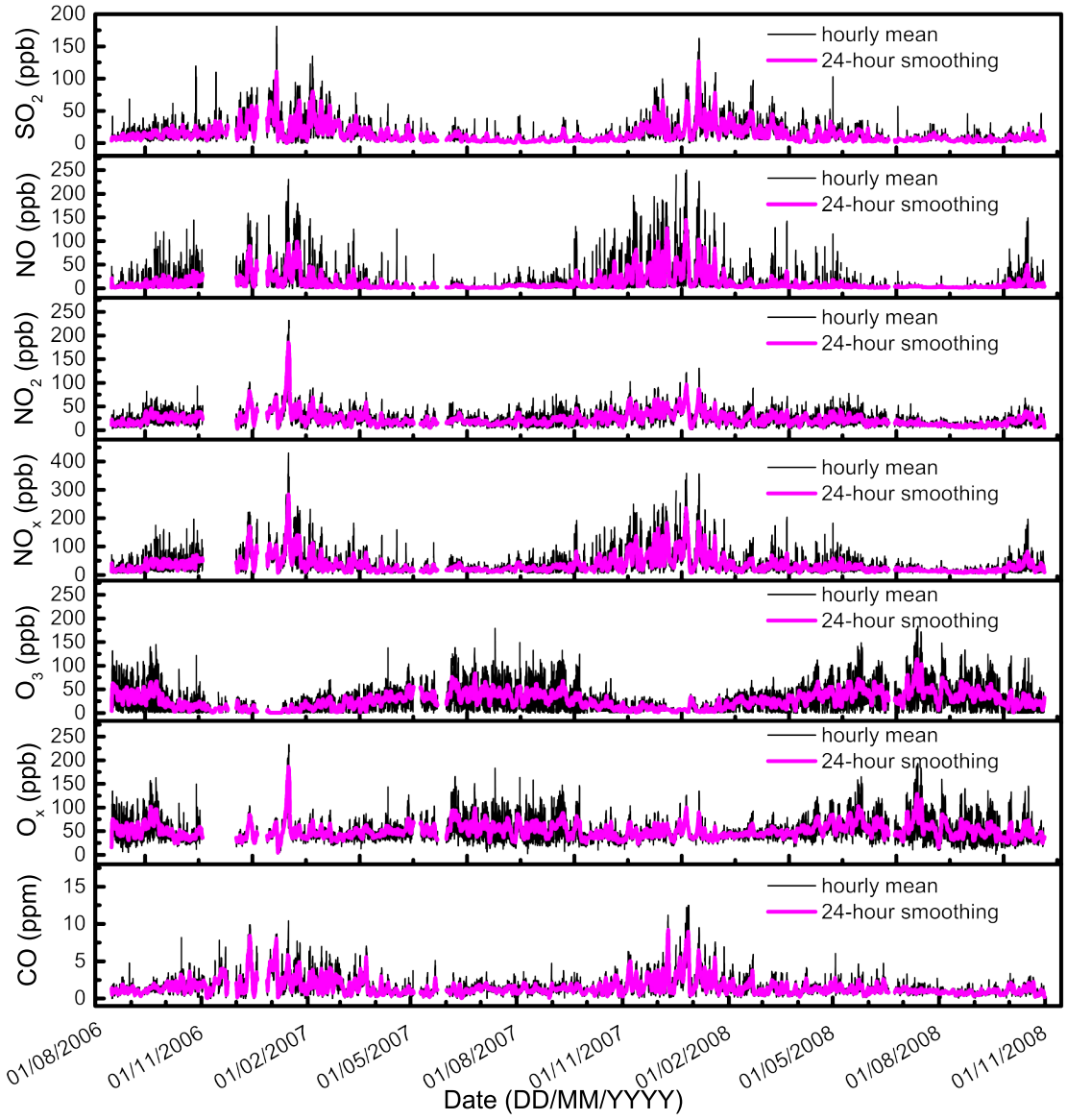
Table 4. The mean net surface flux intensities (i.e. Flux_2007 and Flux_2008), the influx intensities (positive; from the NCP to Yufa; In_2007 and In_2008), the outflux intensities (negative; from Beijing to Yufa; Out_2007 and Out_2008), and the mean concentrations (i.e. Cont._2007 and Cont._2008) during 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



846

847 **Figure 1.** The location information of the Yufa site.



848

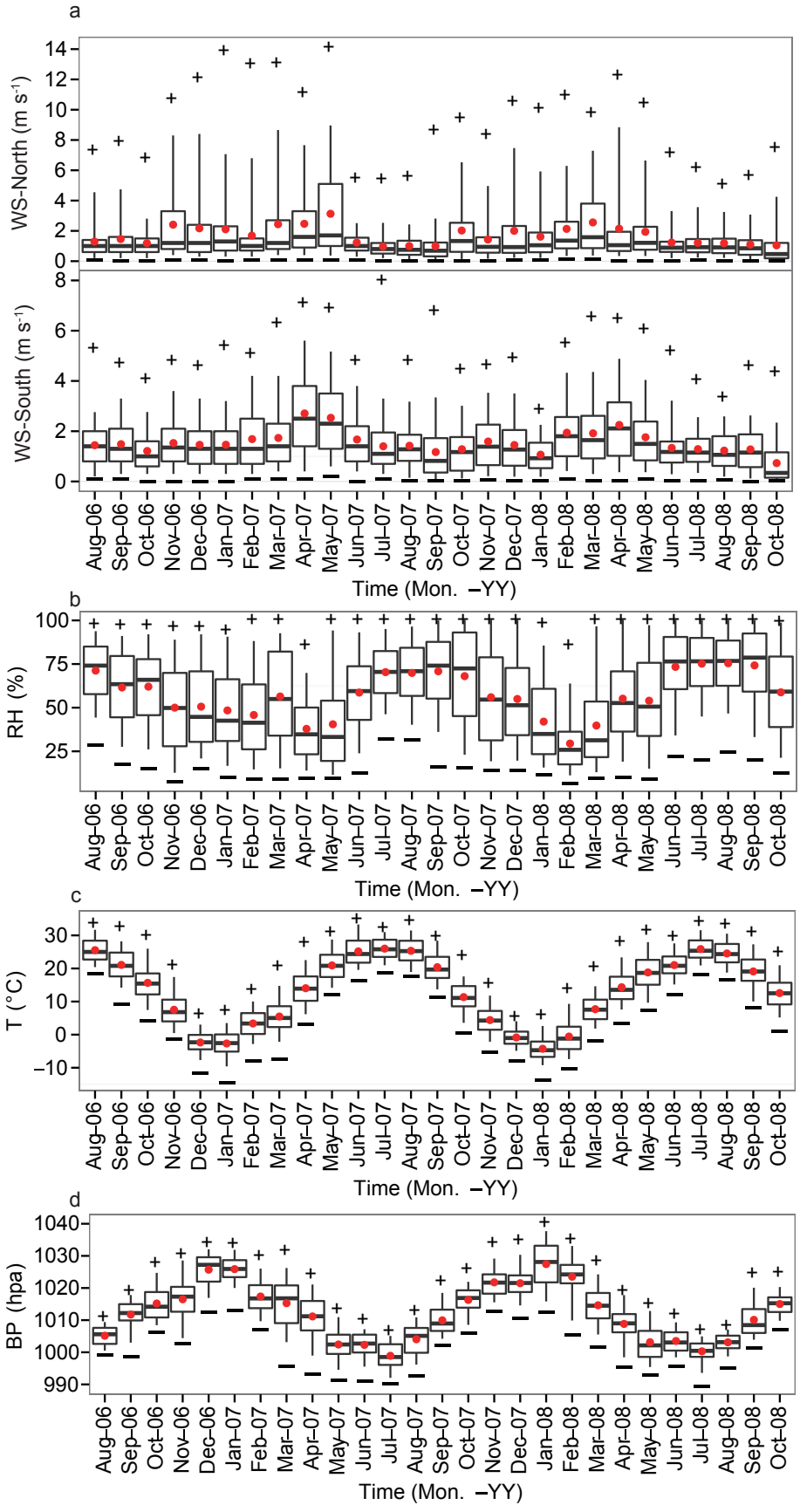
849 **Figure 2.** Time series of hourly mean (black line) and 24-hour smoothing

850 concentrations (red line) of SO_2, NO, NO_2, NO_x, O_3, O_x, and CO at the Yufa site from 15

851 August 2006 to 31 October 2008.

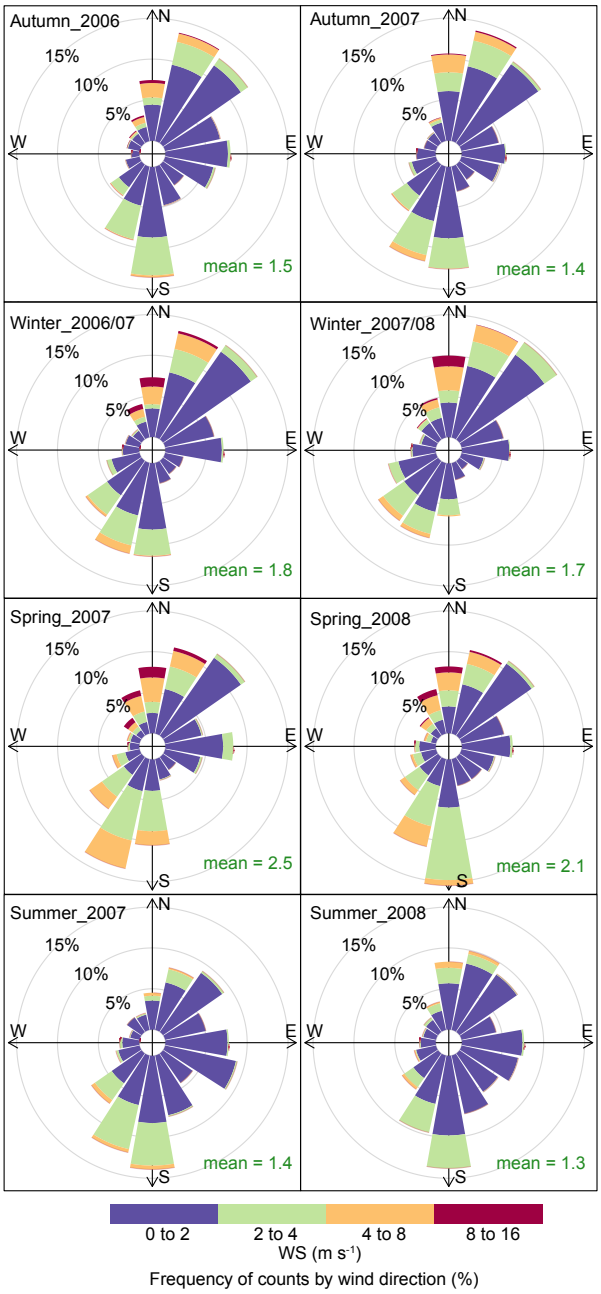
852

853


a
WS-North (m s-1)
WS-South (m s-1)
Time (Mon. –YY)
b
RH (%)
Time (Mon. –YY)
c
T (°C)
Time (Mon. –YY)
d
BP (hpa)
Time (Mon. –YY)
Aug–06
Sep–06
Oct–06
Nov–06
Dec–06
Jan–07
Feb–07
Mar–07
Apr–07
May–07
Jun–07
Jul–07
Aug–07
Sep–07
Oct–07
Nov–07
Dec–07
Jan–08
Feb–08
Mar–08
Apr–08
May–08
Jun–08
Jul–08
Aug–08
Sep–08
Oct–08

854

855 **Figure 3.** Monthly statistics of wind speed (WS) for north wind (a) top and south wind
856 (a) bottom, relative Humidity (RH) (b), temperature (T) (c) and barometric pressure
857 (BP) (d) at the Yufa site. The red point represents the mean value. The black cross bar
858 stands for the median value. The black box and whisker denote the 5th, 25th, 75th
859 and 95th percentiles, respectively. The plus and minus symbols represent the
860 maximum and minimum, respectively. It should be clarified that the North and South
861 wind here is different from the wind direction definition in meteorology. The South
862 wind here is the wind with direction from 90° to 270°, while the North wind is from
863 0° to 90° and from 270° to 360°.
864



865

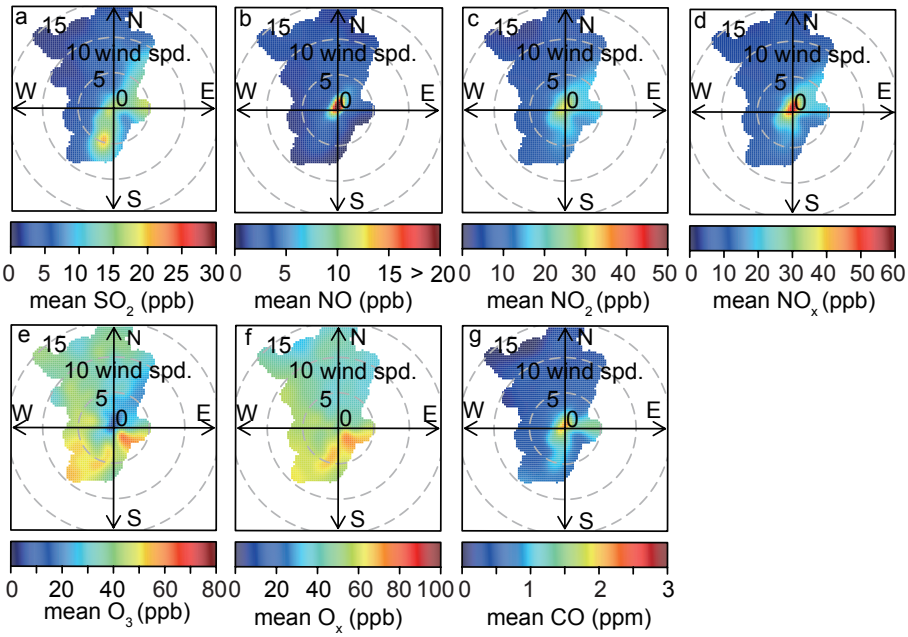
866 **Figure 4.** Wind rose plots based on frequencies of hourly data in Autmun_2006,

867 Autumn_2007, Winter_2006/07, Winter_2007/08, Spring_2007, Spring_2008,

868 Summer_2007, Summer_2008. Spring (MAM): March, April, and May; Summer (JJA):

869 June, July, and August; Autumn (SON): September, October, and November; Winter

870 (DJF): December, January, and February.



871

872 **Figure 5.** Bivariate polar plots for SO_2 (a), NO (b), NO_2 (c), NO_x (d), O_3 (e), O_x (f) and CO

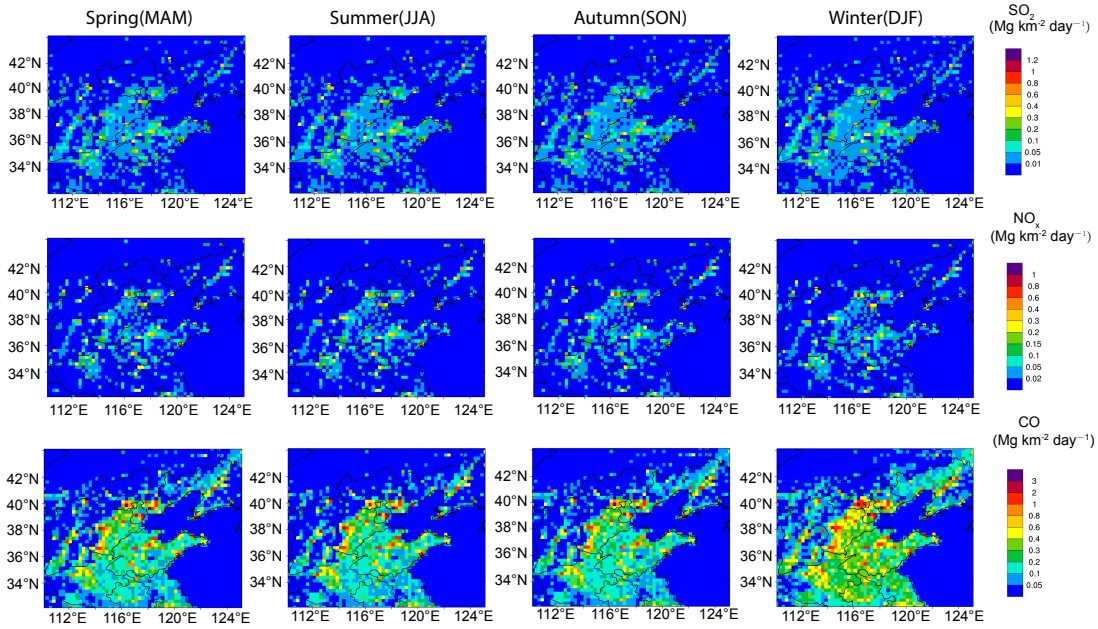
873 (g) concentrations based on hourly average data at the Yufa site from 1 September

874 2006 to 31 August 2008. The colour scale shows the concentrations of pollutants in

875 ppb (or ppm specially for CO) and the radial scale shows the wind speed (m s^{-1}),

876 which increases from the centre of the plot radially outwards.

877

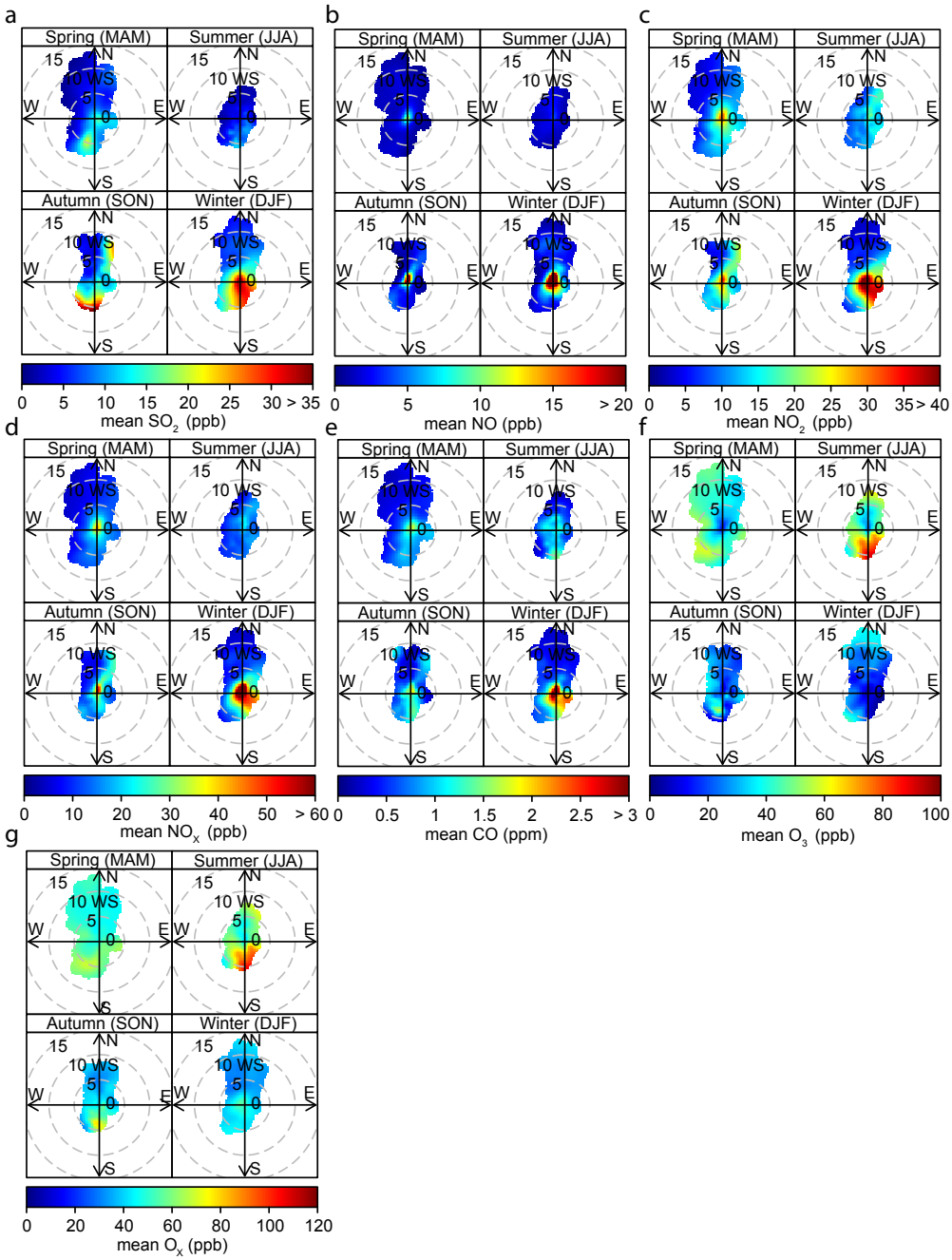


878

879 **Figure 6.** Spatial distribution of seasonal NO_x, CO, and SO_2 emissions in 2008 based

880 on the Multi-resolution Emission Inventory of China (MEIC; www.meicmodel.org)

881 (unit: Mg km^{-2} day^{-1}, horizontal resolution: 25 km×25 km).



882

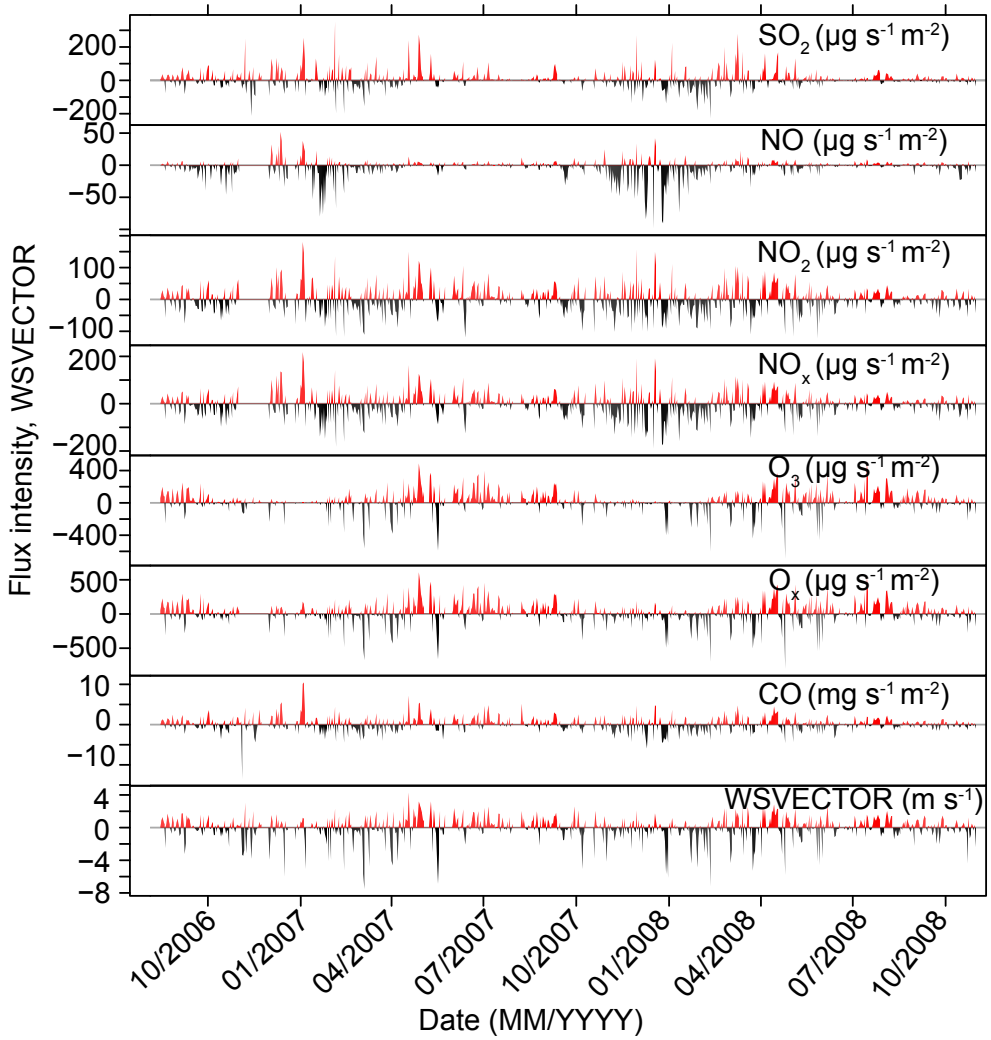
883 **Figure 7.** Seasonal bivariate polar plots for SO_2 (a), NO (b), NO_2 (c), NO_x (d), CO (e), O_3

884 (f), O_x (g) and concentrations based on hourly mean data at the Yufa site from 1

885 September 2006 to 31 August 2008. The colour scale shows the concentrations of

886 pollutants in ppb (or ppm specially for CO) and the radial scale shows the wind speed

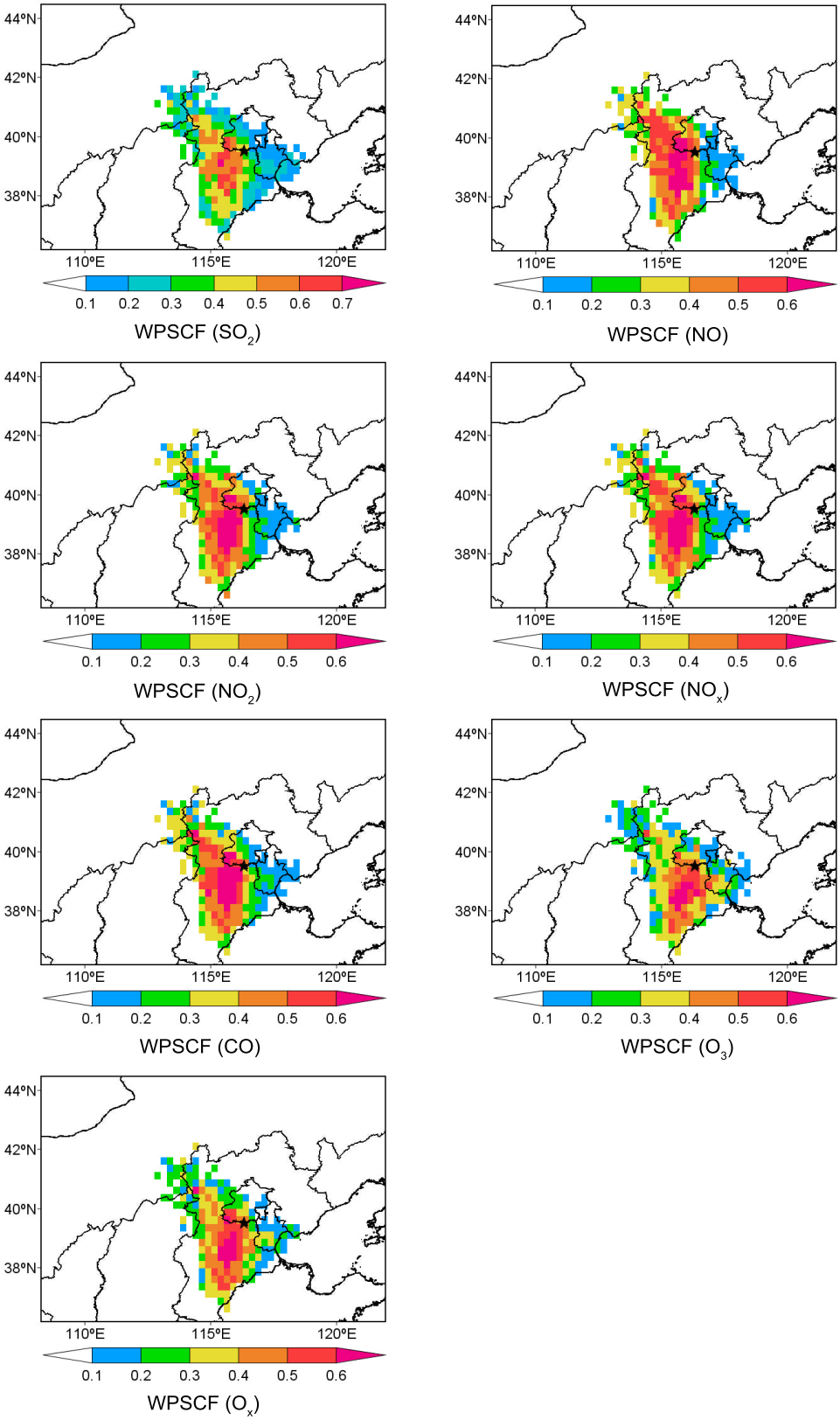
887 (m s^{-1}), which increases from the centre of the plot radially outwards.



888

889 **Figure 8.** Time series of surface flux intensity (i.e. flux per unit cell, μg s^{-1} m^{-2} or mg
890 s^{-1} m^{-2}) for SO_2, NO, NO_2, NO_x, O_3, O_x, CO and wind vector (i.e. WSVECTOR=
891 $-\frac{1}{n}\sum_{j=1}^{n} WS_j \cdot cos\theta_j$, m s^{-1}) based on daily average data at the Yufa site from 15
892 August 2006 to 31 October 2008. The red shaded line indicates the positive transport
893 direction of gaseous pollutants from south to north (i.e. from the NCP to Yufa) and
894 the black shaded line represents the negative transport direction of gaseous
895 pollutants from north to south (i.e. from Beijing to Yufa).

896


44°N
42°N
40°N
38°N
110°E
115°E
120°E
0.1 0.2 0.3 0.4 0.5 0.6 0.7
WPSCF (SO2)
0.1 0.2 0.3 0.4 0.5 0.6
WPSCF (NO)
0.1 0.2 0.3 0.4 0.5 0.6
WPSCF (NO2)
0.1 0.2 0.3 0.4 0.5 0.6
WPSCF (NOx)
0.1 0.2 0.3 0.4 0.5 0.6
WPSCF (CO)
0.1 0.2 0.3 0.4 0.5 0.6
WPSCF (O3)
0.1 0.2 0.3 0.4 0.5 0.6
WPSCF (Ox)

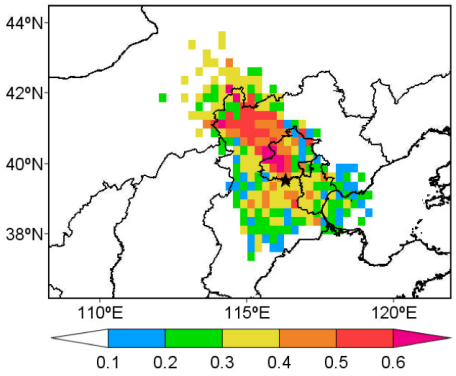
897

898 **Figure 9.** The PSCF maps for the SO_2 (a), NO (b), NO_2 (c), NO_x (d), O_3 (e), O_x (f) and CO

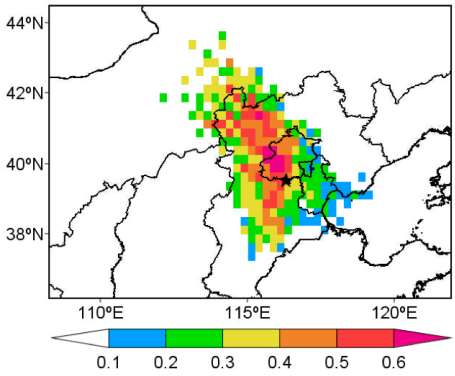
899 (g) surface influx intensity (positive; from the NCP to Yufa). The criterion value of the

900 surface flux intensity is set to greater than the median values, i.e. 20, 3, 30, 34, 40, 94,

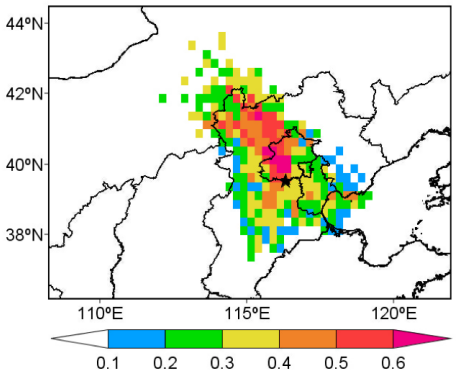
901 and 1200 μg s^{-1} m^{-2} for SO_2, NO, NO_2, NO_x, O_3, O_x, and CO, respectively.



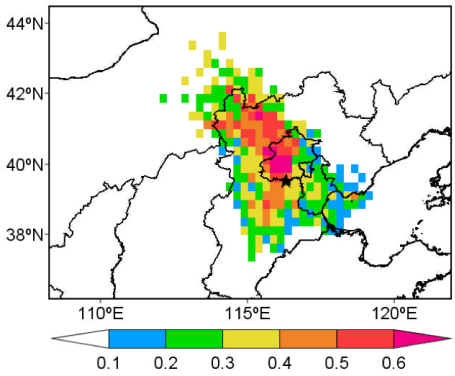
WPSCF (SO_2)



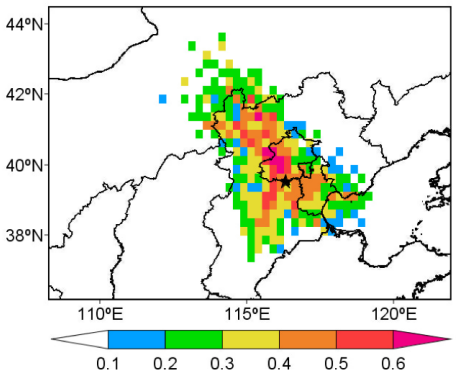
WPSCF (NO)



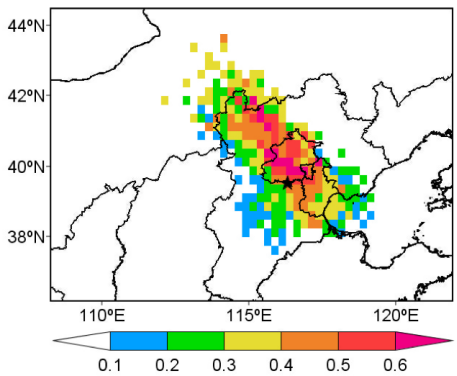
WPSCF (NO_2)



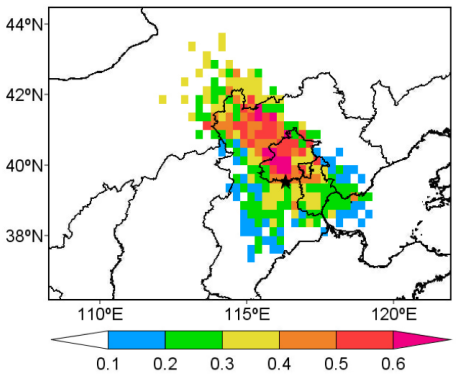
WPSCF (NO_x)



WPSCF (CO)



WPSCF (O_3)



WPSCF (O_x)

902

Figure 10. The PSCF maps for the SO_2 (a), NO (b), NO_2 (c), NO_x (d), O_3 (e), O_x (f) and CO (g) surface outflux intensity (negative; from Beijing to Yufa). The criterion value of the surface flux intensity is set to lower than the median values, i.e. -18, -5, -35, -43, -16, -67, and -1200 μg s^{-1} m^{-2} for SO_2, NO, NO_2, NO_x, O_3, O_x, and CO, respectively.

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

Yingruo Li[1], Chunxiang Ye[1,2], Jun Liu[1], Yi Zhu[1], Junxia Wang[1], Ziqiang Tan[1], Weili Lin[3], Limin Zeng[1], Tong Zhu[1*]

[1]SKL-ESPC and BIC-ESAT, College of Environmental Sciences and Engineering, Peking University, Beijing, 100871, China

[2]Now at School of Chemistry, University of Leeds, Leeds LS2 9JT, UK

[3]Meteorological Observation Center, China Meteorological Administration, Beijing, 100081, China

*Corresponding Author: tzhu@pku.edu.cn

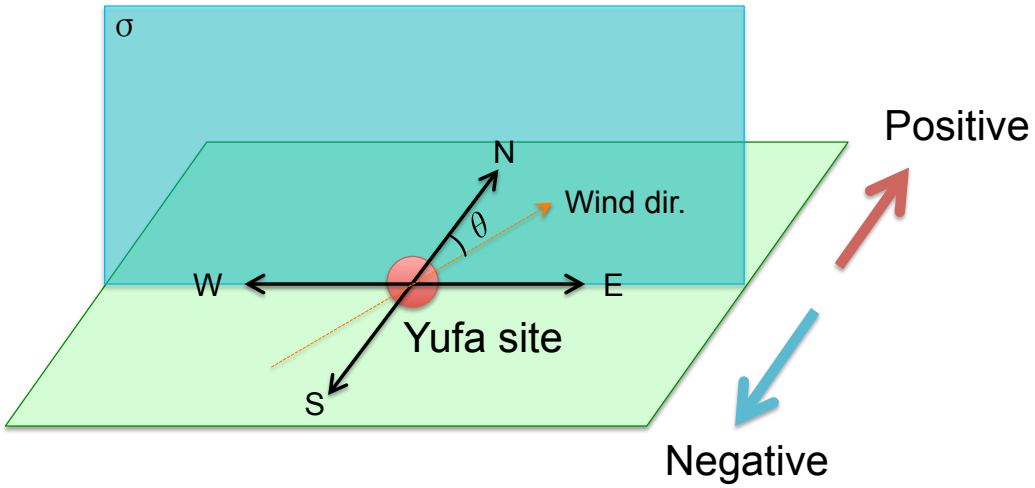


Figure S1. Principles for calculating gaseous pollutant flux: the wind vector along the north-south direction multiplies concentration of gaseous pollutant equals the gas pollutant flux. The direction from south to north (i.e. from the NCP to Beijing) is defined as positive, and vice versa.

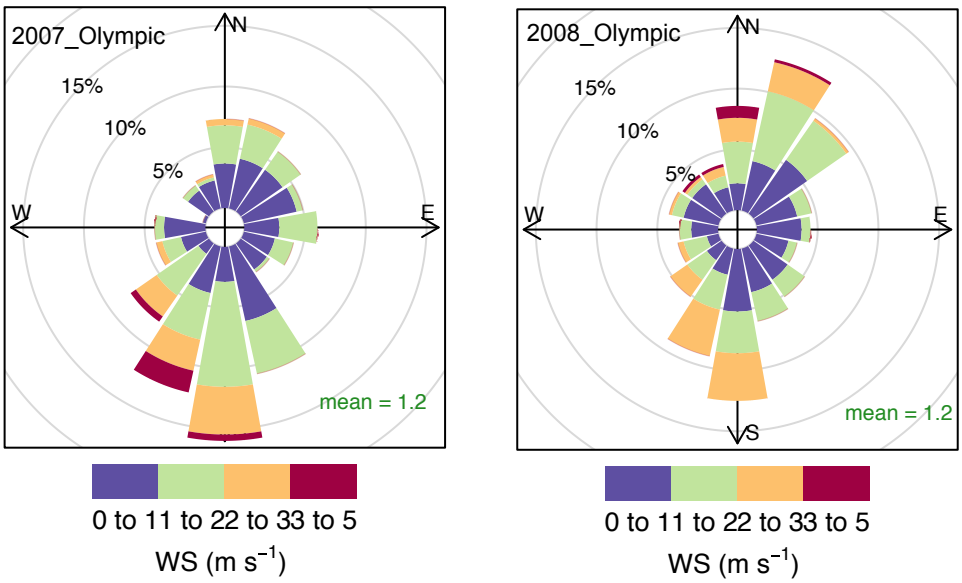


Figure S2. Wind rose plots during the 2008 Beijing Olympic period from 8 August 2008 to 20 September 2008 (2008_Olympic, the right panel) and the same corresponding period of 2007 from 8 August 2007 to 20 September 2007 (2007_Olympic, the left panel) based on hourly wind data.