RC1:

This paper applied the WRF-CMAQ model to simulate the air quality over the Beijing-Tianjin-Hebei area and calculate the trans-boundary fluxes to Beijing, Tianjin, and Shijiazhuang. This paper used a new method that assessed the pollutants transport by vertical surface flux calculation instead of usually used scenario analysis.

Author’s reply: We feel encouraged to receive the reviewer’s recognition for our work. We treasure the valuable comments raised by the referee and have followed them in revising the manuscript. Please check the below for the point-to-point responses.

(1) Using this method, it is easily to understand the pollutants inflows from each direction or each surrounding city to the objective city. But the inflows from one city didn’t necessarily mean those pollutants were from that city. It might be generated from the upflow cities. Did the authors consider about that? And is there any consideration about solving this?

Authors’ reply: We appreciate for the valuable comment. Indeed, the transport flux itself could not tell whether the pollutants are from the neighbor city or from other upstream cities, and this problem is one of the major disadvantages of the flux method. However, the goal of using the flux method is mainly focused on the transport from different directions, rather than the contribution of different cities. By putting together the transport characteristics of different cities as is done in our study (see Fig. R1), we have obtained a general transport feature in the BTH region, which can facilitate a qualitative understanding of where the fluxes are mainly from.

As shown in Fig. R1, in January, the $PM_{2.5}$ that flows into Beijing from Baoding on the southeast mainly happens at a higher level, so the $PM_{2.5}$ may origin from a larger area upstream. Then we can track backward along the dark arrow, and the inflow may come from Baoding, Shijiazhuang or even Xingtai. In July, the transport directions between Baoding and Shijiazhuang are different at the lower and the upper level. We can infer that the inflowing $PM_{2.5}$ into Shijazhuang mainly come from Baoding rather than farther regions to the northeast.

We admit that the flux approach cannot quantitatively evaluate the contribution from each city, and we hope that future studies can combine the flux method with other methods such as tagging models to quantitatively assess the contribution of different cities or regions on the transport pathways identified in the current study.

We have included the preceding discussions on the limitations in the revised manuscript. (Page 15, Line 403-409)

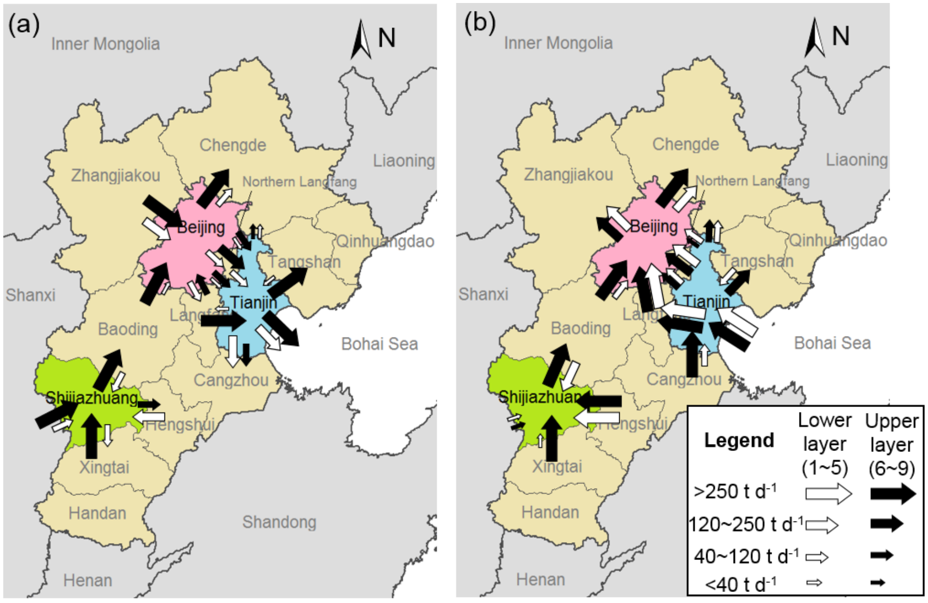

Figure R1 The transport fluxes through each boundary segment of the three target cities in January (a) and July (b). The size of the arrows represents the amount of the fluxes, while white and black arrows denote fluxes at the lower (layer 1-5 in the model, from the ground to about 310 m) and upper (layer 6-9 in the model, from about 310 m to about 1000 m) layers, respectively.

(2) Wind directions are very important to calculate the pollutants fluxes (Figure 3 and Equation 1). But in the model evaluation section (S2), the authors only evaluate the wind speed, temperature, and humidity. I would suggest the authors to evaluate the wind direction in the simulation results.

Authors' reply: We thank the referee for the good suggestion. We evaluated the wind direction using the same method as the other meteorology parameters. The results are shown in Table R1. The bias of wind direction at 10m (WD10) falls within the benchmark, but the gross error exceeds the benchmark for both January and July. The larger gross error is partly caused by the lower precision of the observation data. The WD10 observations only have 16 different values, while the simulation could have any value between 0 and 360. For example, if the real WD10 is 125 degree, the value will be reported as 140 degree. Even if the simulation is exactly 125 degree, an additional gross error of 15 degree will be introduced. In addition, compared to other similar simulation studies in China (e.g., Hu et al., 2016; Zhao et al., 2013), the gross error of WD10 falls in a similar range. Therefore, we still believe that the simulated meteorology field shows a reasonable agreement with the observation.
We have added the data and the discussion above in our SI (Page 3, Line 37 – 50).

Table R1 Comparison of simulated and observed wind direction

Parameter	Index	Unit	Benchmark[a]	Jan-2012	Jul-2012
Wind direction (WD10)	Observation Mean	deg	-	203.9	175.2
	Simulation Mean	deg	-	222.4	174.8
	Bias	deg	≤±10	-2.64	-1.47
	Gross error	deg	≤30	43.23	43.7

a. The benchmarks used in this study are suggested by Emery (2011).

RC2:

This paper analyzed the flux flow between cities in BTH area, northern plain in China, with a commonly used transport model WRF-CMAQ. It is an important issue for policy makers to understand the regional transport of air pollution, and would be helpful in decision of emission control strategy. The paper is clearly written and easy to follow. I suggest its publication when the following issues are further stressed or discussed.

Author's reply: We appreciate the reviewer's valuable comments which help us improve the quality of the manuscript. We have carefully revised the manuscript according to the reviewers' comments. Below is our point-to-point responses to the issues raised by the reviewer.

(1) Language. There are some grammar errors in the manuscript and the language should be polished.

Authors' reply: We sincerely apologize for the deficiency in language. We have gone through the text carefully and corrected the grammar errors.

(2) Lines 56-57, Page 3. It is not quite persuasive, since the non-linear relationship is considered in the DDM and RSM methods.

Authors' reply: We thank the referee for pointing out the problem. The expression in the manuscript was not quite accurate and might cause misunderstanding. We believe that all methods based on chemical transport models, including the DDM and RSM methods, are able to consider the non-linear relationship between emissions and concentrations. However, the sensitivity of $PM_{2.5}$ concentrations to emission perturbation, which DDM and RSM aim to quantify, is different from the inter-city transport of $PM_{2.5}$. Assuming that the emission reduction in the source region leads to a 30% reduction in $PM_{2.5}$ concentration in the target region, the transboundary transport of $PM_{2.5}$ may not be 30% because of the nonlinearity in the emission-concentration relationships.

Zhao et al (2017) found that during winter time, the $PM_{2.5}$ could response negatively

to NOx reduction. Therefore, if we use sensitivity approach such as brute-force method, DDM and RSM to assess the regional transport of $PM_{2.5}$, the result may deviate (probably underestimate) from the real case.
We changed our expression in the manuscript for a better understanding of the insufficiency of the DDM and RSM methods for our study (Page 3, Line 55-57).

(3) Lines 128-129, Page 6. The authors stated that the biases of simulated meteoro logical field and $PM_{2.5}$ concentrations fall in a reasonable range. For meteorological field, the statement could be supported by Table S1, with the evidence by Emery et al., 2001. For $PM_{2.5}$ concentrations, however, we could not think it is "reasonable" as no further information is given. The bias could be quite large in some case, and some major components such as SOC were largely underestimated as indicated by the authors. Therefore, I suggest that the authors provide the evidence or criterion to justify the model performance., or describe the current simulation progress (model performance) in BTH region.

Authors' reply: We thank the referee for the valuable suggestion. We calculated additional indices including Mean Fractional Bias (MFB) and Mean Fractional Error (MFE), and compared them with the benchmark values suggested by Boylan and Russell (2006). The definition of the two indices can also be found in the research of Boylan and Russell (2006). The simulation results of the $PM_{2.5}$ concentration are well within the model performance criteria. We have added the new statistical results as well as the benchmark values in Table 2 in the manuscript.
Due to the limitation of monitoring sites, the benchmark is not applicable for the evaluation of $PM_{2.5}$ components. Therefore, we compare the model performance with other studies in the BTH region. The results are summarized in Table R2. All of the studies underestimate the sulfate concentrations. The underestimation ranges between 9% and 79%, and most of them are larger than 30%. The nitrate simulation results vary in different studies, but the majority of the studies tend to overestimate its concentration. The concentration of EC is usually much lower than the other four components, which may contribute to the large discrepancy in the simulation results in different studies. For OC, although some studies overestimate the concentration, more studies exhibit a lower concentration than observation. Generally speaking, the biases of the $PM_{2.5}$ components in the current study have similar magnitude to other recent studies in the BTH region.
We have included the discussion in the revised SI (Page 7, Line 69-80)

Table R2 Summary of the $PM_{2.5}$ component simulation results for the BTH region in recent studies

Time	Site	SO_4^{2-} NMB (%)	NO_3^- NMB (%)	NH_4^+ NMB (%)	EC NMB (%)	OC NMB (%)	Reference
2005 annual	Tsinghua, Beijing	-14	13	10	-24	-36	Wang et al., 2011
2005 annual	Miyun, Beijing	-36	62	9	-17	-52	Wang et al., 2011
14 Jan – 8 Feb, 2010	Beijing	-72	-32	-5	124	26	Liu et al., 2016
14 Jan – 8 Feb, 2010	Shangdianzi, Beijing	-78	-24	-13	36	-7	Liu et al., 2016
Jan, 2010	Peking university, Beijing	-39	85	33	101	-2	Liu et al., 2016
14 Jan – 8 Feb, 2010	Shijiazhuang, Hebei	-79	-35	-7	81	38	Liu et al., 2016
14 Jan – 8 Feb, 2010	Chengde, Hebei	-78	48	-10	-39	-50	Liu et al., 2016
14 Jan – 8 Feb, 2010	Tianjin	-72	0	9	149	85	Liu et al., 2016
11-15 Jan, 2013	Beijing	~ -73	~ -43	-	-	-	Wang et al., 2014
Jan 2013	Handan, Hebei	-9	33	-11	50	37	Wang et al., 2015
Jul 2013	Handan, Hebei	-32	-3	8	96	30	Wang et al., 2015
Oct – Nov, 2014	7 sites in the BTH region	-48	16	-25	87	-37	Zhao et al., 2017
22 Jul – 23 Aug, 2012	Xiong County, Hebei	-52	95	2	120	-25	This study
22 Jul – 23 Aug, 2012	Ling County, Shandong	-57	79	-14	117	-1	This study

(4) Section 3.2, Page 9. The authors described the difference in flux pattern between Jan and July. However, the reasons for the difference is not further discussed, and the seasonal mechanisms in pollution transport remained unclear. More information should be provided here.

Authors' reply: We appreciate for the valuable comment. What determine the seasonal transport flux are mainly two factors, the wind speed and the $PM_{2.5}$ concentration in the upstream areas. The $PM_{2.5}$ concentration is related to both the meteorology condition and the emission, with the upstream emissions being the most important factor. If we understand the roles of emissions and winds in the transport, we can answer the question of the seasonal mechanisms. Therefore, we combined the response of this comment with the fifth one below.

(5) Related with Q4, the paper described the pattern of pollution transport between cities, which is helpful for policy making. For scientific issue, however, the main factors influencing the transport were not sufficiently discussed. Could the author explain the roles of emissions and meteorological condition on the transport using the cases presented in the paper?

Authors' reply: We combine the response of this comment with the fourth one. To better understand how the wind and concentration affect the transport fluxes, we have made several wind rose plots for different cities, different seasons and different heights. Besides the traditional wind rose plot that displays wind direction with wind speed frequencies, we also made plots that display the wind direction with $PM_{2.5}$ concentration frequencies. We chose the ground layer and the 7th layer to represent the lower layer and the upper layer respectively. The plots for Beijing are shown in Fig. R2 as an example. The plots for the other two cities are displayed in the SI (Fig. S3 and Fig. S4).

In January, the dominant wind directions near the ground ranges from northwest to northeast. The NNE wind has the highest frequency, while the NW wind has the highest wind speed (Fig. R2(a)). The dominant northern winds reflect the winter monsoon. Although the concentration coming with the northern winds are relatively low because of the low emission rate on that direction (Fig. R2(b)), the high frequency and wind speed also cause an overall strong transport from the northwest to the southeast. Wind directions and the corresponding concentrations are quite different at the upper layers (Fig. R2(c), (d)). The prevalent northern wind remains (though the dominant directions shift slightly from NNE to NW), and the frequency of southwestern winds is much higher than that at lower layers. Moreover, the $PM_{2.5}$ concentrations that come with southwestern winds are much higher than the other directions. The strong emission in southern Hebei (which lies on the southwest direction of Beijing), especially the elevated sources may be responsible for the high concentration from the southwest. Therefore, in January, the dominant northwestern winds account for the Northwest-Southeast pathway at both lower layers and upper layers, while the large emissions on the southwest direction mainly caused the Southwest-Northeast pathway at upper layers.

In July, the dominant wind directions at the lower layer are the southeastern directions, reflecting the summer monsoon (Fig R2(e)), and coincidentally the highest concentrations also come along with the southeastern winds (Fig R2(f)). Emissions from Tianjin, Langfang, and Tangshan may influence Beijing by the southeastern winds. The emission and the wind direction both contribute to the Southeast-Northwest pathway at the lower layers. The high frequency wind directions shift clockwise to the southern directions at the upper layers in July, as is shown in Fig. R2(g), and the southwest wind and the southeast wind are both important. Moreover, the directions with high concentrations also shift to both the southwest and the southeast directions (Fig. R2(h)). Therefore, in July, the dominant southeastern winds and the emissions on the southeast directions caused the Southeast-Northwest pathway at both the upper and the lower layers. The Southwest-Northeast pathway is a combination result from the southern winds and the emissions, which is different from that in January.

Similar analysis can be made to the plots for the other two cities. Due to the length limitation, we put the plots into the SI. The plots in Fig. R2 and the discussions above has been included in our revised manuscript (Page 10-11, Line 262-287).

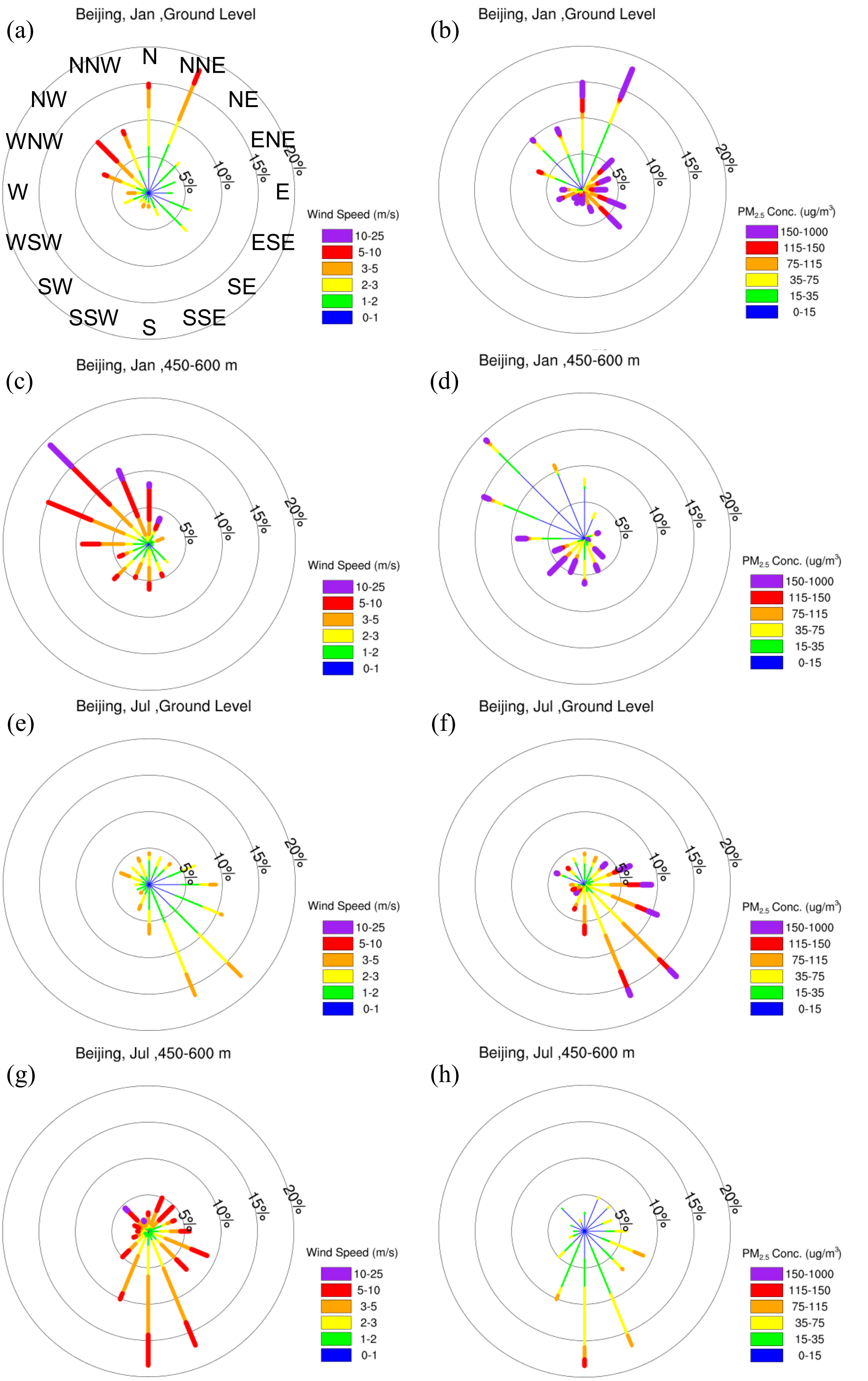

Figure R2 The wind rose plots showing the frequency of wind speed (a, c, e, g) and $PM_{2.5}$

concentration (b, d, f, h) at different wind directions for Beijing. The ground level and the 7th level (about 450-600 m) in the model are chosen to represent lower levels and upper levels. The percentages denote the frequency.

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