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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Contributions of pollutants from North China Plain to surface ozone at theShangdianzi GAW station" *by* W. Lin et al.

W. Lin et al.

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—We thank both referees for their very constructive comments and suggestions. We revised our manuscript according to their comments and suggestions.

Response to comments by referee 1
Anonymous Referee 1
This manuscript of Lin et al. reports 3-years records of ozone and related trace gases measured at Shangdianzi station in the North China. The data should be very important for the scientific community because rare of works have been done in this polluted region. The paper is written in a compact format, overall structure is good, and figures are clear and well produced. However, the referee has a major concern on the methodology that used to address the main topic, i.e. the regional contribution of ozone

from the North China Plain. The paper can be published in ACP, if this major concern and other comments were appropriately addressed. The major concern: The authors use the surface measured winds to classify the ozone data into two contrast parts: natural background and polluted background, and further try to quantify the regional contribution from the North China Plain. This method has a large uncertainty because the surface winds at complex topography are not appropriated to address long-range transport issue. The Fig.1 shows that the site locates in a valley with a lot of mountains surrounded; therefore the wind should be large affected by local topography, and by the thermal driven mesoscale processes, like mountain valley breezes, which can be clearly seen from Fig 2.

—We understand the concern about the topographic influences on the surface winds and would like to carefully address this issue. On the one hand, we agree that the surface winds at Shangdianzi (SDZ) may be influenced to a certain extent by the local topography. On the other hand, we believe that the surface winds there are more influenced by meso- to large-scale airflows.

— We analyzed the annual and seasonal average diurnal variations of surface wind vectors at SDZ and three sites in the North China Plain, Beijing Observatory (BJO), Gucheng (GCH), and Bazhou (BZ). The wind vectors were calculated from the surface wind data from 2004 to 2007 (from 2006 to 2007 for GCH). The results show that in all seasons except winter, the wind direction at SDZ changed nearly clockwise, with southerly winds around noon and northerly winds around midnight. Similar clockwise circulations of wind exist at the other three sites. This similarity in wind leads to similar wind roses of these sites. The major differences in the wind roses between SDZ and the other sites are the frequencies of northwesterly and southeasterly winds. The valley topography around SDZ and the trend of the valley reduce the frequencies of these winds. Since both SDZ and the sites in the North China Plain show common features of wind rose, i,e., northeasterly and southwesterly prevailing winds, it is likely that the winds

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observed at SDZ contain more information of meso- to large-scale airflows than small-scale influences of the local topography.

So the classified results based on surface winds can only represent a relative small scale but not a regional scale as large as 1000 x 1000 km2 that defined by the author. In another word, the results here can probably only reveal the impact from Beijing urban area, which is just located in the southwest of the site about 100km. However, the referee strongly suggests that author addressing the regional contribution from the North China Plain and comparing it with that from Beijing. Back trajectory analysis could be an appropriate methodology, or a method combining large scale wind data (like NCEP or ECMWF reanalysis) with the local winds could also be a choice. The referee find that the back trajectory analysis has been adopted in a sister paper using the same dataset by Meng et al., (ACPD, 8, 9405-9433, 2008).

— The largest dimension of the North China Plain is about 500 km, so the 1000 x 1000 km2 is considered to be a region with SDZ in the middle. Since Beijing is the largest city in the North China Plain and its urban area is a large population center nearest to SDZ, the impact from this area is surely most important to the results at SDZ. On the other hand, the air pollution in Beijing is caused not only by local emissions but also by transport of pollutants from surrounding areas, particularly the areas southwest to southeast of Beijing. Recent studies (An et al., 2007; Kang et al., 2006; Meng et al., 2006; Ren et al., 1004; Yan et al., 2005; Zhang et al., 2004) suggest that transport of pollutants from the surrounding areas contribute significantly to the pollution in Beijing and it can make about half of the pollutants concentrations in Beijing under unfavorable conditions. This means that air masses transported from Beijing to SDZ carry a certain portion of pollutants from outside of Beijing, particularly south of Beijing. Therefore, regional contribution from the North China Plain cannot be neglected although the influence of Beijing urban area is very important (this is emphasized in the revised manuscript).

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— To further support our view, we have also made use of NCEP/NCAR reanalysis data. The seasonal mean surface vector winds indicate that westerly to southerly surface winds dominate in North China in spring, summer, and autumn and northwesterly winds in winter, suggesting that transport of pollutants from southwest to SDZ is often a regional phenomenon, particularly during warmer period of the year. We also compared the mean surface and 925 hPa vector winds for warmer months (April-October). Both surface and 925 hPa winds are southwesterly, favoring the transport of air masses from the North China Plain to SDZ. More detailed and typical patterns showing regional transport over the North China Plain can been seen in Fig. 15 of the recent paper by An et al. (2007).

— Back trajectory analysis had been done, but the results are not adopted in this paper. Some details of this analysis are included in the sister paper (Meng et al., 2008). The results from trajectory analysis suggest that air masses from the south are relatively more polluted than from the north, consistent with the result from surface wind analysis.

— In summary, the surface wind observed at SDZ seemed to be more influenced by meso- to large-scale airflows than by the local topography. This probably has something do with the trend of the valley, which is nearly parallel to the prevailing winds in the north of the North China Plain. The ideal location of SDZ, i.e, at the northeast edge of the North China Plain, combining with ideal trend of the valley near SDZ, makes it possible to quantify the influence of the North China Plain on surface ozone at SDZ according to ozone and wind measured in the surface layer.

— In the revised paper, a paragraph is added to Section 3.3 at the end and readers are referred to this interactive comment. In addition, a file containing figures will be sent to the editor and then to the referees.

In fact, if only using the local wind data some results presented in the current paper seem to be duplicated. For example, the seasonal difference of ozone between SW and NE has also been shown in Fig.7. The patterns of Figs. 10 and 11 are similar with Fig 3, maybe because the winds have strong diurnal variation with SW in the afternoon and NE in early morning and thereafter the difference between SW and NE somewhat reflects the range of O3 diurnal change in Fig.3.

— Figure 6 in our manuscript show overall average distributions of O3 and Ox in different wind directions, while Fig. 7 the seasonal difference in the distributions. The winter (DJF) distribution is completely different from those of other season. Although the curves in Figs. 10, 11, and 3 look similar, these three figures do show different data. The difference in ozone concentration between SW and NE winds is related to the diurnal and seasonal variations of wind direction. On average, the winds have strong diurnal variation with SW in the afternoon and NE in early morning and seasonal variation with SW mainly in summer and NE mainly in winter. The statistical results of both diurnal and seasonal variations are needed.

Other minor points: Section 3.1 contains too much discussion comparing the observed results with those from a remote mountainous site Waliguan, which is totally different site from the Shangdianzi. It should be better to make some comparison with previous works done in this area and also other similar polluted region in North Hemisphere, like Northeast US etc.

— The section is intent to explain the difference of daily ozone pattern at a regional background station close to city with that at a global background station. Some comparison are added in the revised version and more comparisons can be see in the sister paper of Meng et al.(2008).

Section 3.2 including Fig. 5 is not useful since it only show a common frequency distribution of ozone. The referee suggests deleting this part or making if for different seasons and putting it in Section 3.1 when discusssing the seasonal differences.

— The seasonal differences are discussed in the revised version now.

Response to comments by referee 2
Anonymous Referee 2
A very large economic growth took place in China during the last decade and nitrogen dioxide emissions strongly increased in this region as documented by satellite observations. This development suggests a simultaneous increase in regional ozone concentration in the planetary boundary layer but only a limited number of studies (referenced in the submitted paper) about tropospheric ozone in China have been published in the reviewed literature. The study presents measurements of ozone and primary pollutants from a surface site (Shangdianzi) approximately 100 km north to Beijing often being in the outflow region of the Beijing area ("North China plain") and the paper provides a convincing interpretation of the measurements including advection regimes. Because the measurements are expected to continue in future providing valuable information of the future development of photooxidant air pollution in the region the reviewer recommends publication in ACP if the following comments are properly taken into account.

Abstract: 1)Line 9, p. 9140: The reviewer suggests to remove "natural (clean)", compare comment 13;

— accepted.

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2)Line 17/18, p. 9140: The last sentence of the Abstract might be changed, possibly by: The emissions of nitrogen oxide in the North China plain cause a decrease in ozone concentrations in winter.

— accepted.

Introduction:

3)Line25, p. 9140: Regarding the strong increase in ozone over Europe you might refer to J. Staehelin, J. Thudium, R. Bühler, A. Volz-Thomas and W. Graber: Surface ozone trends at Arosa (Switzerland), Atmos. Environ., 28, 75-87 (1994);

— accepted.

4)Line 9, p. 9141:"and etc." should be removed;

— accepted.

5)Line 15, p. 9141 and line 26, p. 9151: Staehelin instead of Staehlin;

— Sorry for misspelling.

6)Line 6, p. 9142: Remove "natural", see comment 13. Site and Observations:

— accepted.

7)Line 22, p. 9142: It should be mentioned, that the measuring principle used for measurements of NO_2 at the site suffers from interference of other NO_y compounds such as PAN and HNO_3. The authors might reference the paper of M. Steinbacher, C. Zellweger, B. Schwarzenbach, S. Bugmann, B. Buchmann, C. Ordonez, A.S.H.

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Prevot, and C. Hueglin, Nitrogen oxide measurements at rural sites in Switzerland: Bias of conventional measurement technique, J. Geophys. Res., Vol. 112, D11307, doi:10.1029/2006JD00791, 2007. This implies that the measured NO2 concentrations have to be viewed as an upper limit, particularly in aged air masses such as in summer;

— Agreed.

8) Measurements run under the program of Global Atmospheric Watch (GAW) of World Meteorological Organization (WMO) are expected to be published at the respective World data center, which should be done prior to publication of the measurements in a reviewed paper. Have the authors already submitted the measurements of ozone of the Shagdianzi station to the World Data Center of Greenhouse Gases?

—Not yet. Submission of these data and those from other GAW station in China subjects to the regulations of CMA. We hope that the submission can start in the following year. As staff of the data owner, we are allowed to publish results obtained using the data.

Results and Discussion:

9) Figure 3: It appears that the curves of the diurnal variation of ozone are not correct at midnight for the January and February. Are the respective "jumps" at midnight an artifact of the used software program? This should be corrected in the revised version;

— The curves are not necessarily to be smooth because they are average diurnal variations in different month. The way of drawing these curves led to the jumps. In the revised figure breaks are inserted between the diurnal curves.

10) Line 23, p. 9144: "ozone and related gases": SO2 is not directly related to ozone (not a precursor), therefore the term "related" should be replaced by "primary

pollutants"; **— accepted.**

11) Fig. 4: The diurnal variation of SO_2 concentrations shows a distinctly different pattern than the other primary pollutants particularly compared to NOx; NOx peaks during the time when inversions usually occur, in line with the expectation of dominant emissions into the regional planetary boundary layer. SO_2 concentrations peak at 16.00 (average over the year) or 11 (in summer) when the PBL is expected to be mixed with that are larger in the layer above the inversion. This might suggest that larger SO_2 concentrations are transported to the measuring site in a layer above the inversion. Please comment on the possible origin of SO_2 and possible transport mechanisms;

—The major SO_2 emissions are from industry, power plants, and domestic heating in Beijing and other cities in the North China plain. A similar pattern of diurnal variation of SO_2 was also found in a rural background site, 110 km south to Beijing city. The data do suggest higher SO_2 concentrations in a layer above the inversion. This phenomenon may be caused by high chimneys in factories and power plants. Since no detailed information about emitters is available and no vertical profile of SO_2 has been measured at the rural sites, there is no conclusive explanation.

12) Table 2: The reviewer does not completely understand what "maximum average" and "minimum average" exactly means and how the numbers are determined. Please clarify;

— The average values for 16 wind direction sectors were determined. The maximum average means the maximum value among the sectors, and the minimum average means the minimum value among the sectors. These are clarified in the revised version.

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13) Line 4-24, p. 9147: The reviewer thinks, that it is rather difficult to justify that ozone concentrations advected by the N-NNE sector to the measuring site represent "natural background ozone" (only) attributable to the transport from the stratosphere and ozone production from natural emissions, because tropospheric ozone increased on a hemispheric (at least intercontinental) scale since World War II. However, it is justified to assume that the difference between the two sectors are caused by the ozone production in the North China plain, which is the most important message in the paper (comp. comments 1 and 6);

— accepted. "Clean" is used instead of "natural" where necessary.

14) Line 13, p. 9148: It is not completely clear, how the "daily accumulated contribution of 240 ppb.hr" is calculated. Please specify;

— It is an integration of the daily average of the difference (10 ppb) over 24 hours (10 ppb x 24 hr = 240 ppb.hr).

15) Line 25, p. 9148: The reviewer suggests to replace: "Because of inactivity of photochemical production in winter" by "Photochemical ozone production in the Planetary Boundary layer is very small in winter and therefore higher NOx concentration in the SW sector leads to chemical ozone destruction (gas titration, i.e. reaction of NO with O3) explaining lower ozone concentration in the polluted sector than in the regional background air".

— Accepted.

Conclusions:
16) Line 7-8, p. 9149: Sentence starting with "Influenced by ..." could be changed to "Surface ozone concentrations at SDZ show some unique properties because of its

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location and the surrounding topography";

— Accepted.

17) Comment to line 15, p. 9149: In order to verify the interpretation of the SDZ measurements, the reviewer recommends additional ozone (and possibly precursor measurements) South East and close to Miyun. During days of strong photochemical activity one might expect that ozone peaking values occur earlier in the day at this site than at SDZ station, similar as reported in A.S.H. Prévôt, J. Staehelin, G.L. Kok, R.D. Schillawski, B. Neininger, T. Staffelbach, and A. Neftel: The Milan phootoxidant plume, J. geophys. Res., 102, 23,375-23,388 (1997).

— Agreed. Yes, Diurnal variations of O3 observed at Miyun in June and July 2006 by Wang et al. (2008) showed that ozone peaking appeared at about 16:00 in June and about 17:00 in July, which was earlier than that in SDZ (ozone peaking appeared at about 18:00 in June and July, 2006). Results in Wang et al. (2008) are cited in Section 3.1 in the revised paper.

References

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