Dear Editor,

We greatly thank the reviewers for their detailed review. Many valuable comments and suggestions were provided, for which we are grateful. Point-by-point responses addressing all the comments were uploaded (and also attached to this file). The manuscript has been revised and improved accordingly.

Best Regards

Chunsheng Zhao

Answers to Referee #1's comments and suggestions:

1) ***In 2.4.1, the dry deposition velocity of SO_2 was taken from Tsi (2010), it should be described if it was suitable to NCP area.***

   Thank you for the suggestion. The following few sentences were added to the revised manuscript to describe that it is suitable to use the dry deposition velocity from Tsai et al. (2010) in our work: "The dry deposition velocity in the NCP is ranges from 0.2-0.8 cm s^{-1}, showing no significant seasonal variations (Pan et al., 2013), which conforms well with the result of Tsai et al. (2010). Thus, it is believed to be appropriate to use the diurnal pattern measured by Tsai et al. (2010) for the NCP region."

2) ***In 2.4.2, the radiation data were not described.***

   Thanks for your comment. The NCAR MM2.4 model is coupled with a Tropospheric Ultraviolet and Visible Radiation (TUV) model, which calculates the photolysis rates needed for photolytic reactions. The radiative properties of the 1st Jul 2009, 1st Oct 2009 and 1st Jan 2010 were modelled to represent the summer, autumn and winter cases, respectively. This information was added to the text in 2.4.2.

3) ***For gaseous oxidation, the VOCs data taken in summer season were used in three seasons. A sensitivity test of VOCs should be done, since it could be a non-linear process.***

   This is a very good point. According to your suggestion, we performed a sensitivity test with doubled/tripled summertime VOCs concentrations while reducing the isoprene concentrations to a half/a third of its summertime values. Results are shown in the revised Table 4. The increased VOCs concentrations during autumn and winter will lead to increased SO_2 oxidized amounts, however, the relative increase of group 2 and group 3 to that of group 1 decreases only slightly with increasing VOCs concentrations. Overall, the VOCs concentrations have little influence in the inter-comparison between the three groups.

Table 1 Simulated SO_2 gaseous oxidation amount for the three different SO_2 diurnal variation patterns, the occurrence frequency (of the three groups) weighting averaged value and the relative increase compared to the nighttime peak case. Case1, Case2 and Case3 respectively represent cases using summertime VOCs concentrations, doubled summertime VOCs concentrations with half of the

summertime isoprene concentrations and tripled summertime VOCs concentrations with a third of the summertime isoprene concentrations.

Case	SO_2 gaseous oxidized amount (ppbv day^{-1}) [Increase relative to nighttime peak case (%)]						
	summer	autumn			winter		
	Case1	Case1	Case2	Case3	Case1	Case2	Case3
Nighttime	2.3	3.1	3.7	4.0	3.0	4.0	4.5
Noontime	2.6 (15%)	3.9 (26%)	4.6 (23%)	4.8 (21%)	3.7 (23%)	4.7 (18%)	5.2 (16%)
Noontime+Nighttime	2.9 (28%)	4.3 (39%)	5.1 (36%)	5.4 (35%)	4.5 (50%)	5.9 (49%)	6.7 (49%)
Weighting Average	2.5 (9%)	3.8 (21%)	4.5 (19%)	4.7 (19%)	3.7 (23%)	4.8 (21%)	5.4 (20%)

4) In 2.4.3, the haze process had not been described.

The aqueous oxidation process of SO_2 in clouds are believed to be more important than that in haze, due to the large liquid water content of clouds. During noontime, aerosol liquid water content (Bian et al., 2014) is especially low, hence contributing far less to SO_2 aqueous oxidation than clouds. The aqueous oxidation process of SO_2 in haze is more complicated than that in clouds, it is highly influenced by the aerosol composition, liquid water content and pH value. The haze-SO_2 interaction is studied therefore in another work of ours and will not be further discussed in this study.

5) In page 5662 line 4-7, the uncertainties were discussed for assuming the trace gas concentration. The conclusion was that will not have influences in the intercomparison between the groups. In fact, this conclusion could be suitable to other two processes, if there were linear. There is a suggestion that the uncertainty discussion should be done in all of three processes.

Thank you for this valuable comment. The uncertainty for the gaseous oxidation process was added according to comment 3.

6) In Fig.5, there are more lines and dots with different colors, which could be identified in electronic version. For paper publication, it would be difficult to identify. Maybe a large figure should be shown in paper publication.

Thank you for this suggestion. Since the printed paper publications will also be in colour, we did not change the figure. However, we will suggest the editorial office to make the figure larger in the ACP version, to make the lines easier to identify.

7) In page 5662 line 17-18, the language is not clear.

To make this clearer, the sentence "At SDZ, high SO_2 concentrations in all but the winter season occur during noontime (Fig. 2a)." was rephrased as "At SDZ, high SO_2 concentrations occur during noontime in all seasons, except for the winter season (Fig. 2a)."

8) For abbreviation sem (in page5657 line 21 and page 5663 line 4) it's better to write in capitals SEM.

Thank you for pointing this out, the abbreviations were changed accordingly to capital letters in our revised manuscript.

Reference:

Bian, Y. X., Zhao, C. S., Ma, N., Chen, J., and Xu, W. Y.: A study of aerosol liquid water content based on hygroscopicity measurements at high relative humidity in the North China Plain, Atmos. Chem. Phys. Discuss., 14, 4089-4118, 10.5194/acpd-14-4089-2014, 2014.

Pan, Y. P., Wang, Y. S., Tang, G. Q., and Wu, D.: Spatial distribution and temporal variations of atmospheric sulfur deposition in Northern China: insights into the potential acidification risks, Atmos. Chem. Phys., 13, 1675-1688, 10.5194/acp-13-1675-2013, 2013.

Tsai, J.-L., Chen, C.-L., Tsuang, B.-J., Kuo, P.-H., Tseng, K.-H., Hsu, T.-F., Sheu, B.-H., Liu, C.-P., and Hsueh, M.-T.: Observation of SO_2 dry deposition velocity at a high elevation flux tower over an evergreen broadleaf forest in Central Taiwan, Atmospheric Environment, 44, 1011-1019, 10.1016/j.atmosenv.2009.12.022, 2010.

Answers to Referee #2's comments:

1) ***The noontime SO_2 peaks have been already reported by several previous works in Beijing (e.g. Wang et al., 2006; Gao et al., 2013) and other locations in China (e.g. Ding et al., 2013). It looks that there may be some difference in rural and urban sites. In urban sites, SO_2 pattern generally have morning and evening peaks because of influence of local emission. This referee suggest that the authors make a throughout comparison on results at the two different type of sites.***

   We thank the referee for the valuable suggestion. We added further discussions on the noontime SO_2 peak phenomenon reported in past studies into the introduction part. Additionally, in the summary section we concluded that the different occurrence frequencies of noontime peak phenomenon and the different contribution of each process to each site are responsible for the distinct noontime peak occurrence times in the long-term averaged diurnal profiles at the different types of sites.

2) ***The authors mentioned about the Mountains Yan and Taihang, the North China Plain and mountain breezes, it will be better to add a topographical map in Figure 1, in which clearly demonstrate the geographical location of the study regions.***

   Thank you for this suggestion. Accordingly, we added several contour lines showing the topographical height of the terrain in the North China Plain and the location of the Taihang and Yan Mountains are marked in the revised Figure 1 (see below).

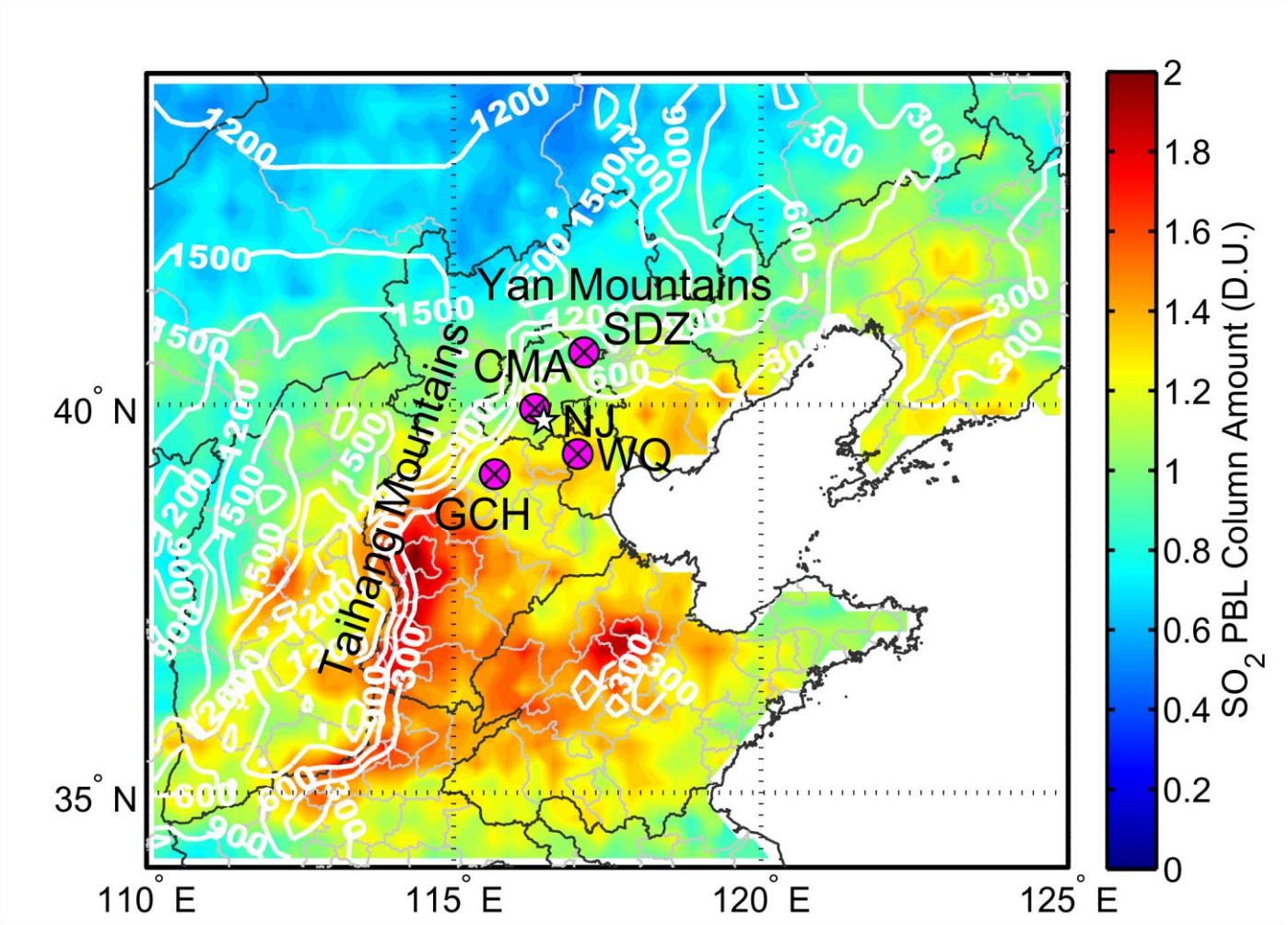

Figure 1 Location of the Shangdianzi (SDZ), China Meteorological Administration (CMA), Gucheng (GCH), Wuqing (WQ) site, the Nanjiao meteorological station (NJ) and the Yan and Taihang Mountains. The average distribution of OMI SO2 column concentration in 2009 is displayed as the shaded contour, while the white contour lines show the terrain height (m).

3) ***Figure 2: Were these plots made using monthly averaged diurnal profiles? If not, what is the data resolution in the y-axis (Month or weekly)? It looks that there are some strange bands, like synoptically scale variations, in these figures. How about using the normalized results to make these plots here?***

Figure 2 is plotted with 14-days moving averaged diurnal profiles, it aims mainly to display the season-diurnal variations of SO_2 concentrations. Synoptic scale variations should have been eliminated through the 14 days moving averaging process. The normalized diurnal variations are already shown in Figure 3, therefore we would like to keep Figure 2 as it is.

4) ***Page 12: On the discussion of stack height (10-240 m), plume rise, an important fact for elevated sources, should also be mentioned here. The effective height of plumes could be much higher than the stack height.***

This is a very good point, thank you for pointing it out. We have added this into section 3.2, on the discussion of stack and plume heights.

5) ***Page 14, Line 16-19. Besides transpiration process, atmospheric turbulence mixing around noontime will also increase the dry deposition. The stronger turbulence mixing will cause more deposition to the surface.***

Thank you for this valuable comment. The sentence was accordingly revised as:
"For surfaces covered with vegetation, dry deposition processes are typically most dynamic during noontime due to transpiration and the strong turbulent mixing processes (Tsai et al., 2010;Raymond et al., 2004), thus noontime SO_2 peaks may create more acid deposition than common nighttime peak variation patterns."

6) ***Sect. 3.3: About the discussion of SO_2 oxidation, because the authors' analysis already suggested that the downward mixing was an important factor influencing the noontime peak of SO_2, its will be better to using some data above the ground surface (e.g. vertical profile in the PBL) to estimate the O_3 concentration for the model calculation.***

This is a good suggestion, unfortunately, the MM2.4 model is a 0-dimensional box model, which does not consider the vertical profile of trace gas concentrations. In past studies, O_3 concentrations were either well mixed or showed slight increases with height within the PBL (Chen et al., 2009;Dickerson et al., 2007;Chan et al., 2004;Geng et al., 2009), thus we believe that our assumptions will not have too much influence on our final conclusions.

Reference:

Chan, C. Y., Zheng, X. D., Chan, L. Y., Cui, H., Ginn, E. W. L., Leung, Y. K., Lam, H. M., Zheng, Y. G., Qin, Y., Zhao, C. S., Wang, T., Blake, D. R., and Li, Y. S.: Vertical profile and origin of wintertime tropospheric ozone over China during the PEACE-A period, Journal of Geophysical Research: Atmospheres, 109, D23S06, 10.1029/2004JD004581, 2004.

Chen, Y., Zhao, C., Zhang, Q., Deng, Z., Huang, M., and Ma, X.: Aircraft study of Mountain Chimney Effect of Beijing, China, Journal of Geophysical Research: Atmospheres, 114, D08306, 10.1029/2008JD010610, 2009.

Dickerson, R. R., Li, C., Li, Z., Marufu, L. T., Stehr, J. W., McClure, B., Krotkov, N., Chen, H., Wang, P., Xia, X., Ban, X., Gong, F., Yuan, J., and Yang, J.: Aircraft observations of dust and pollutants over northeast China: Insight into the meteorological mechanisms of transport, J. Geophys. Res., 112, D24S90, 10.1029/2007JD008999, 2007.

Geng, F., Zhang, Q., Tie, X., Huang, M., Ma, X., Deng, Z., Yu, Q., Quan, J., and Zhao, C.: Aircraft measurements of O3, NOx, CO, VOCs, and SO2 in the Yangtze River Delta region, Atmospheric Environment, 43, 584-593, http://dx.doi.org/10.1016/j.atmosenv.2008.10.021, 2009.

Raymond, H. A., Yi, S.-M., Moumen, N., Han, Y., and Holsen, T. M.: Quantifying the dry deposition of reactive nitrogen and sulfur containing species in remote areas using a surrogate surface analysis approach, Atmospheric Environment, 38, 2687-2697, http://dx.doi.org/10.1016/j.atmosenv.2004.02.011, 2004.

Tsai, J.-L., Chen, C.-L., Tsuang, B.-J., Kuo, P.-H., Tseng, K.-H., Hsu, T.-F., Sheu, B.-H., Liu, C.-P., and Hsueh, M.-T.: Observation of SO_2 dry deposition velocity at a high elevation flux tower over an evergreen broadleaf forest in Central Taiwan, Atmospheric Environment, 44, 1011-1019, 10.1016/j.atmosenv.2009.12.022, 2010.

Answer to W. Zhou's suggestion:

W. Zhou: The authors present an interesting analysis of noontime peak SO_2 in the northern China plain. The authors are suggested to discuss if SO_2 was consumed by lower clouds via aqueous processing when SO_2 peaks occurred. Previously, SO_2 in Texas power plant plumes was rapidly lost via aqueous processing of scatters clouds, the corresponding SO_2 lifetime of which was 2~3 hours (Zhou et al., 2012).

Thank you very much for this valuable comment. We are aware that the aqueous chemistry in lower clouds can lead to rapid loss of SO_2 within the boundary layer. In Sect. 3.3 of our paper, we already have some discussions on the impact of heterogeneous chemistry on SO_2 scavenging. Since past studies have not shown clear diurnal variations in cloud coverage or cloud liquid water content (LWC), we assumed a constant LWC throughout the day. A more detailed study on SO_2-cloud interaction will be shown in our future work. Again, we greatly appreciate your suggestion, which is very valuable for our future work.