Response to referee #1 (acpd-11-C15278-2012)

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We thank the referee for her/his careful review and valuable comments on the manuscript. Below is our point-by-point response to the questions. The reviewer's comments are outlined in italic type and our responses are in regular type.

General comments:

In this work, the authors investigated the potential impacts of regional atmospheric circulation patterns, which are determined via obliquely rotated T-mode principal component analysis (PCA) of surface layer pressure over North China, on regional transport pathways and air quality over Beijing. The authors found that CT 1 (high to the west with a strong pressure gradient), which is characterized by a northwestern origin, and CT 6 (high to the northwest), which has air mass sources mostly from the north and east, are the two favorable CTs for good air quality in Beijing. And they believed that CTs are the primary drivers of day-to-day variations in pollutant concentrations over Beijing and its vicinity. I have some questions which are needed to be addressed.

1. How about the contributions of local emissions and air pollutants from surround regions to the air quality in Beijing? How do CTs impact on Beijing air quality? Via reducing local pollutants concentration or transporting air pollutants to Beijing?

Reply: Beijing is located on the northwest border of the North China Plain (NCP), which is one of the most polluted places in the world. Beijing is surrounded by the Yanshan Mountain in the west, north and northeast (Fig. 1). The local emissions sources for air pollution include power supplies, domestic heating, and industrial, vehicular, and biogenic sources. Regional transport can be important sources of atmospheric particles and gaseous pollutants. Dust storms are one of the major non-Beijing natural sources in spring, and their frequency occurrence decreased significantly in recent years. Apart from dust storm period, the non-Beijing sources (NBS) are mainly from the south and east-south places (An et al., 2007; Wang et al., 2011; Wu et al., 2011; Chen et al., 2007). The local anthropogenic sources are dominated in Beijing during the episode with a northerly transport path. More than 20% of the ground SO_2 mixing ratio in Beijing was estimated to be transported from external sources (Yan et al., 2005). Mobile measurement suggests that the transport fluxes of primary air pollutants to Beijing increase significantly during the episode with a southerly transport path (Wang et al., 2011; Wang et al., 2009). During sustained wind flow from the south, Hebei Province can contribute 50–70% of Beijing's $PM_{2.5}$ mass and 20–30% of ozone productions (Streets et al., 2007). The maximum NBS contribution was estimated to be 60–80% when Beijing encountered heavy PM_{10} pollution episode (An et al., 2007; Chen et al., 2007).

The impact of CT on air quality is via both changing local pollutants concentration and transporting air pollutants to Beijing. From the source to the monitoring sites, the concentration levels of the observed air pollutants are controlled by the emission strengths and the physical (dispersion, transport, coagulation, wet/dry deposition) and chemical processes along the transport paths. These processes in determining the concentrations of the air pollutants rely on the circulation patterns. The local meteorological variables and boundary-layer and regional transport characteristics that affect air quality are both strongly controlled by the synoptic-scale circulation. Our analysis show the CTs were characterized with distinct local meteorology and air mass origins, and therefore different CT is associated with different air pollutant concentration. For example, CT 1 has strong northerly wind and high mixing height that favor the dispersing of local emitted pollutants. Meanwhile, the frequent northerly transport paths in CT 1 are usually related to very low contributions of non-Beijing sources.

2. What are the major differences between northern air mass and southern air mass and their impacts on Beijing air quality? Dust storm, which usually comes with northern air mass, can impact Beijing air quality and PM10 concentration. The impact of dust storm on Beijing air quality is expected to be associated with certain atmospheric circulation patterns. But I do not find this information in CTs analysis.

Reply: As the regional distributions of emission sources (details are shown in Zhang et al. (2009)) and topography, the northern air mass is often in favor of dispersion and associated with less transported anthropogenic pollutants from the surroundings, while persistent southern air mass is usually characterized with high ratio of external pollutants and polluted air quality in Beijing. Besides, dry and cold air masses in Beijing are usually from northern regions, and southerly air masses are frequently moist and warm.

Severe sandstorms are generally related to Siberian anticyclones or Mongolian cyclones, and depend on soil moisture content over the source regions (Chinese and Mongolian deserts). Dust storms are one of the major non-Beijing natural sources only in spring, and their frequency occurrence decreased significantly in recent years (Zhu et al., 2008). The frequency of dust storms that lead to high PM_{10} concentration at surface of Beijing is very low, especially for the period 2005-2009. About only 3% of very bad visibility (< 2km; 7.2% occurrence of total hours) in Beijing during

1999-2007 are associated with dust (Zhang et al., 2010). Except for dust storm period, the external PM_{10} sources are mainly from the south and east-south places (i.e. Hebei province, Tianjin and Shandong). Figure N7 shows that good visibility (> 20km) in Beijing is associated with northerly, northwesterly and westerly wind greater than $3ms^{-1}$, suggesting that less transported pollutants (or PM_{10}) from northwestern and northern regions with respect to long-term mean (1999-2007). As a result, the impact of dust storm on PM_{10} concentration is not obviously associated with a certain circulation during 2005-2009. But, we think dust storm has partly contributed to the mean PM_{10} loadings of those CTs with fast northern air masses (e.g. CT 1).

Special comments:

1. *P33466, L3: "provided holistic evaluation". It is hard to say the evaluation in this study is holistic.*

Reply：Accepted. The word "holistic" is replaced with "quantitative".

2. *P33466, L9-16. It seems the authors believed that regional transport pathways associated with the 9 CTs show more significant impacts on Beijing air quality rather than local meteorology. How do the authors think about the impacts of local meteorology associated with the 9 CTs on Beijing air quality?*

Reply：We believe there is some misunderstanding. Our result show that the synoptic circulation influencing air quality is mainly via local meteorology and regional transport. The statistical analyses show that the circulation pattern has distinct local meteorology and plume transport pattern. The local air quality depends on both local meteorology and regional transport, which are both controlled by circulation pattern. CT 9 displayed a relatively low wind speed and high frequent of south wind at Beijing, which in turn resulted in stagnant air parcels and limited dispersal of pollutants. The fast hygroscopic growth of aerosol due to high relative humidity in CT 9 may significantly increase aerosol mass extinction efficiency and reduce the visibility. The more discussion about the impact of local meteorology on air quality has been presented in the manuscript.

3. *P33467, L1: "e.g. frequent precipitation". Based on table 1, precipitation day frequency associated with the 9 CTs has little impact on air quality. CT 8 and 9 have high precipitation day frequency, but they still have bad visibility.*

Reply：The influence of precipitation on visibility has two folds. One is the positive effect of wet deposition; the other is negative effect of increasing relative humidity that could severely degrade visibility (e.g. fog and haze). Since CTs 5, 8 and 9 have continuous pollutants transported from the south, the reduction of air pollutants via wet deposition is also limited. The dependence of visibility in Beijing on relative humidity and wind direction and speed are shown in Figs N7&N6 (Zhang et al., 2010). Their result shows a clear relation of visibility degradation to southerly wind. The high concentration of air pollutants and fast hygroscopic growth of aerosol due to high relative humidity under CT 5, 8 and 9 could significantly degrade the visibility. Moreover, visibility reduction is also caused by high humidity (fog), air pollutants, or combination of both; it is nature that high frequency of precipitation is to certain level associated with bad visibility.

4. *P33467, L3-6. The relative contribution of synoptic circulation to SO2 can be high up to 41±36%, two times higher than other species. Why?*

Reply: The contribution is calculated by the ratio of the CT driven the anomaly to the total anomaly of SO_2 concentration during the Olympics. There are three reasons for this high value: (1) The total anomaly of SO_2 concentration during the Olympics (about –20%) is not large compared with other species; (2) SO_2 is a species have a relative long life time than other pollutants, such as NO_x, and can be transported long distance. Beijing has much lower SO_2 emission than its southern regions, therefore circulation plays an important role in control of SO_2 concentrations in Beijing; (3) The types 5 and 7 with high SO_2 mixing ratio didn't occur, while the frequency of Type 6 with lowest SO_2 concentration doubled during the Olympics. Therefore, the circulation-driven anomaly could explain larger ratio of the total anomaly of SO_2 concentration. It should be noted that the uncertainty of this evaluation is also larger than other species.

5. P33470, L14-15. Why is the domain determined from 32 to 49 N and 103 to 129 E? Can the region of the domain be changed? If the domain is changed, how about the results associated with CTs? Will the conclusion be changed? Sensitive study should be presented here

Reply: The grid numbers (27 × 18) has the same size as to that of "Western Mediterranean" domain (17°W to 09°E and 31°N to 48°N) by Philipp et al. (2010). The region (32 to 49 N and 103 to 129 E) is good representative of synoptic scale, and Beijing is roughly at the center. Since the good spatial and temporal stability of T-mode PCA for weather classification has been presented in detailed comparisons by Huth et al (2008; 1996), here we just briefly discussed the result of sensitivity study. We changed the predefined domain a slightly (a degree southward) and found very similar results of circulation classification (Fig. SN1). The main correlations of CTs and air quality (visibility as an example) are not changed (Fig. SN2). The best air quality is associated with “high to west-north” (CT 1), while CTs “unique” and “high to east” are related to poor air quality.

We agree with the reviewer that stability of circulation classification (i.e. the degree of similarity of groupings based on temporal subsamples of data and on a slightly different grid; Huth 2008) could be an important issue. We have discussed it in the revised manuscript.

6. P33470, L16-18. Why do the authors not include 06:00 UTC (14:00 LT) reanalysis data to represent daily circulation type? The radiosonde coverage shows little relation to surface layer pressure which is used to determine CTs in this study. If the authors use the 06:00 UTC or 0000+06:00 UTC reanalysis data, what are the major changes to the conclusions? It should be discussed in the manuscript.

Reply: While we appreciate your suggestions, we still prefer 08:00 LT reanalysis data to present the CT because that the more radiosonde data could significantly improve the accuracy of simulated meteorology both in upper-level and surface while the advanced data assimilation techniques used in NCEP reanalysis. Xie et al. (2012, J. Geophys. Res., submitted) shows that there are strong correlations (covariance calculated by using the ensemble perturbations) between the sea level pressure and 850 hPa height at the observed sounding (shown in Fig. N5). Especially, the improvement in sea level pressure is much larger after assimilating the entire sounding observation than that of only assimilating the sea level pressure at surface. Moreover, the results of classification are easily applied for predicting air pollution in synoptic perspective since the 08:00 LT synoptic charts of SLP are widely used as the daily weather pattern in China. The averaged SLP map for 00:00 and 06:00 UTC can not represent the real circulation pattern, which is useful for understanding the physical processes; it has not been used for circulation classification yet. Therefore, we think 00:00+06:00 UTC reanalysis data is not suitable for circulation classification.

The sensitivity study is performed using 06:00 UTC SLP data. Because the total sequence of input data has changed, the comparative result shows some differences between the CTs although some patterns are very similar (Fig. SN3). However, the main conclusion is not changed: the good visibility is associated with “high to west-north with large pressure gradient”, and CT “high to east” is related to poor air quality (Fig. SN4).

7. P33470, L19-22. Beijing is a megacity, while Beijing Capital International Airport is located at suburbs about 20-30 km away from downtown. How can the meteorology parameters measured at the rural site to represent the local meteorology characteristics at the urban region?

Reply: Actually, the BCIA site was almost been surrounded by the urban region (Fig. 1b in the ACPD manuscript). The more developed urbanization around the BCIA can be found in updated Google earth maps. The PKU and BCIA are both in the north of Beijing urban region, which has a distance about 24 km. The long-term (10 years) meteorological data are only available at WMO 54511 site and BCIA in Beijing. The 54511 site, locating in the south of Beijing, has the similar surroundings but low temporal resolution data (3h interval). Further, we compared BCIA and PKU data, and the difference is minor. Consequently, we think the meteorological measurements at BCIA can represent the local characteristics in Beijing.

8. P33471, section 2.2. Here, the authors only used one site air pollutants measurements to represent Beijing air quality. It is a problem. How to evaluate the representativeness of PKU site?

Reply: Agree. The PKU site is located on the campus of Peking University in the northwest of Beijing city, about 400m north of the fourth ring road, 5 km west of the Olympic Park, and 10 km from the center of Beijing. The campus is a primarily residential and commercial area without industrial sources. Wehner et al. (2008) suggested that an examination of the spatial variability of $PM_{2.5}$ mass and chemical composition in 1999–2002 showed only minor differences between the PKU site and a downtown site. Therefore, PKU site is assumed to be representative of a typical urban environment in Beijing (Cheng et al., 2008; Wehner et al., 2008; Wang et al., 2010; Chou et al., 2011). Even though one site may not reflect the average air pollution levels in Beijing, the temporal variation of the air pollution at this site is the same as that in whole Beijing, hence the association of this variation with circulation pattern is the same for the whole city.

9. P33474, L25-27. The modeling results from WRF are very important for FLEXPART and footprints simulations and related discussions. The validation of WRF results is too weak in this paper. Statistical analysis of measurements and simulations is better than just say "were generally in good agreement with the observations". One site validation is not enough. Many WMO sites can provide basic measurements of meteorology parameters. Synoptic chart can also be used to evaluate the wind field and precipitation field. Planetary boundary layer height and wind field are important for air pollutant transport and trajectories simulation. Therefore, PBLH and vertical wind field should be validated too.

Reply: Accepted. We thought the averaged value of a variable from one grid of WRF output represent an area average of 4km×4km, which is not agree with in-situ site observation. This is one of the potential reasons for the disagreement. We present additional evaluation of the model output with the surface temperature, humidity, wind speed and wind direction of other two sites (shown in Figs. S2b-d) as well as vertically measurements from radiosonde data (Figs. S21-24). Meanwhile, the statistical bias analyses are shown. Moreover, the GPS-based water vapor at PKU station is used to evaluate the WRF-simulated column-integrated precipitable water vapor (shown in Fig. S2d). (The synoptic chart is not used for evaluation of WRF model because it is difficult to be quantified.) For the purpose of dynamical downscaling, we hypothesize that such utilization in the mesoscale model can generate realistic regional structures that can not be resolved by the coarse-resolution forcing data (i.e. NCEP FNL data). Beijing and its vicinity do have complex topography and land uses, and therefore complicated air circulations. Consequently, the synoptic chart based on NCEP data is not used for evaluation of WRF output wind field.

The PBL height is an important parameter characterizing the structure of lower troposphere. Unfortunately, we did not have direct measurement data of PBL height. However, Wang et al. (2011) has validated the simulated PBL height by WRF model, which is configured with the same initial conditions (.e.g. MODIS land use data and input meteorology) and physics parameterizations (options) to ours. They compared hourly averaged WRF-predicted PBL in urban Beijing with the temporal variations of aerosol extinction coefficient retrieved from lidar measurements (Supplementary Figs. S3 and S4 of acp-11-11631-2011; http://www.atmos-chem-phys.net/11/11631/2011/acp-11-11631-2011-supplement.pdf). The results show that the calculated PBL heights were generally in agreement with the top boundary of the extinction coefficients. Therefore, we believe the WRF modeling for this study could have the same performance of calculating the PBL height.

10. P33478, L10. Why does CT 3 have good visibility (11.1 km)? CT 3 has low wind speed (2.08 m/s), low PBLH (1.14 km), high RH (65.7%) and most local sources. All these are not favorable for good visibility.

Reply: The CT 3 is characterized with northern wind (Fig. 4) and north-plume-transport pattern (Fig. 5). The prevalent origins from north indicate relatively clear air masses, which is favorable for good visibility. The dependence of visibility in Beijing on wind direction and speed was presented in figure N7 (Zhang et al., 2010). Their result shows the clear relations of good visibility to the north wind.

11. P33481, L17-19. CT 3 has heavy PM10 and BC loading, but its AOD are not very high. Why?

Reply: The transport pathways and local wind speed of CT3 both indicate frequent stable conditions. In this background, it will significantly elevate aerosol concentration (e.g. PM10 and BC) at low level. At the upper level, the northerly transport pathways indicate clean air at upper level. Moreover, CT 3 has lower PBL height. Therefore, the total column AOD is is not very high in CT3.

12. P33482, L10-12. Particles also have large variations of the emission rate. Why the distributions (variances) of the particle concentration in some specific CT are not as large as those of the gaseous concentration?

Reply: This is a good question and we need further study. We think there might be three reasons. One is that the gaseous pollutants have larger seasonal variations; the second is that chemical processes (transformation) are more important for gaseous species. Besides primary particles from the direct emission, secondary aerosol account a large and even major portion of PM in Beijing, the formation of secondary particles have much less spatial variation than primary particles.

13. P33482, L14-15. According to the authors' previous explanation, CT 1 can trans- port northern clean air to reduce local air pollutant concentrations in Beijing. But we found SO2 concentration is very high under CT 1. Fossil fuel combustion for heating can also enhance BC emission, why is BC loading still very low under CT 1?

Reply: SO_2 was mainly from coal burning, while BC could come from coal and biomass burning, which usually happened in the southern region of Beijing. Traffic emission is another major BC source that has small seasonal variation. As a result, the concentration of SO_2 has larger difference between heating and non-heating period (shown in Fig. N8) than that of BC loadings. Therefore, the frequent very high concentration of SO_2 in winter could significantly elevate the averaged value in CT 1. Moreover, less removal processes and lower chemical transformation rate due to the cold and dry airmass and less precipitation in winter could also contribute to the high SO2 concentration under CT1 in winter. CT 1 has low BC loading because of the good dispersion conditions, higher PBL height and clean air masses.

14. P33483, L8-10. Did the authors analyze the relations between CTs and sulfate concentration? How about the variations of ammonium and nitrate during the same period? They are major components of particle pollutant in Beijing, thus the authors should discuss about them. How about O3 during the same period?

Reply: Since the sulfate observation is only available in the period from 24^{th} July to 20^{th} September, the relations between CTs and sulfate concentration were not analyzed from a long-term perspective. According to the Fig. 12, some associations can be drawn. The polluted episodes that characterized with high PM_{10} and sulfate concentrations are persistently dominated by CTs 5, 8 and 9. The increase of sulfate loading is closely related to the southerly transport pathways. On contrast, CTs 1, 2 and 6 are associated with the low sulfate aerosol loadings. The decrease of sulfate aerosol is mainly due to fast north wind and wet scavenging via precipitation. But, these associations may need to be validated using a long-term data.

Ammonium nitrate is in thermal equilibrium with ammonia and nitric acid, temperature also plays an important role in determining the concentrations in particulate ammonium nitrate, this complicates the analysis. The nitrate and ammonium aerosol data are not accessible to us. But, the temporal variations of chemical compositions in PM_1 mass loading at PKU during the emission restriction period can be found in Huang et al (2010). They concluded that the percent contribution of nitrate and ammonium elevated significantly with total PM_1 concentration. The time series of hourly O_3 at PKU in the same period are shown in (Chou et al., 2011). Compared to the pollution episodes, the O_3 concentration decreased during the Olympics.

15. P33483, L12-17. According to Fig. 10, CT 5 and 9 are associated with high SO2 concentration, while CT 6 and 8 are associated with low SO2 concentration. From Aug 24th to 30th, the dominate CT is 5 and 9, but SO2 concentration is always very low. Why?

Reply: The low SO_2 mixing ratio in late August is due to the fast transformation of primary emitted SO_2. SO_2 is chemically oxidized to sulfate by both gas and liquid phase processing. The concurrent measurements of SO_2 and sulfate show major airborne sulfur pollutants (Fig. 12). Additionally, the temporal variation of gaseous H_2SO4 can be found in Zheng et al (2011). The concentration of sulfate (H_2SO4) was very high while SO_2 value is very low during 24-30th August. It suggests the more efficiently oxidizing and removing of the primary emitted SO_2, and indicates that either much stronger sulfate production exists at the SO_2 source region or other sulfate production mechanisms are responsible for the sulfate production (Zheng et al., 2011). Moreover, higher boundary height and lower residential coal consumption in the summer also contribute to this.

16. P33483, L24-29. The authors said "under emission control the air quality improved significantly during the Olympics". Now the authors concluded that "The large variations in air pollutant concentrations and the delay in air quality improvement cannot be explained by the control measures only because the pollutant episodes and clean Olympics episode were characterized by different circulation types". How do the authors comment on the contribution of emission control to air quality improvement during the Olympics?

Reply: Thanks, the sentence has been revised. Our quantitative analyses suggest that both emission control and CT contribute to the improvement of air quality during the Olympic. Air quality can vary in response to changes in pollutant emission and weather conditions. For the whole period of emission control (20 July – 20 September), the low background and mean concentrations of air pollutants in and around Beijing can be mainly attributed to the decreased emission sources. But with respect to short-period (e.g. day-to-day) variations, the key driving force is the change of meteorological conditions (spanned) from local to synoptic scale. Apparently, severe pollution episodes, the delay in air quality improvement and large variations of day-to-day air quality during the emission control are not closely related to the variation of emissions. These day-to-day variations of air pollutants, however, can be mostly explained by the variation of CTs and their associated local meteorology (e.g. precipitation) and regional dispersion and transport conditions. The pollution episodes were persistently dominated by CTs 5, 8 and 9, and had more air masses with southern origins and shorter transport distances; during the Olympics, circulation types favorable for dispersion and wet scavenging contributed significantly to the improvement in air quality. Our results suggest the anomaly of CT could partly explain the reduction of air pollutant during the Olympics. Therefore, the clean air quality during the Olympics is due to the combination effects of emission reduction and favorable weather conditions.

Evaluation of the potential effects of these unprecedented control measures on air quality in Beijing provides valuable information for furthering both scientific understanding and future policy.

17. P33485, L10- P33486, L13. I think the quantitative analysis of the impacts of synoptic circulation and emission reduction on air quality during the Beijing Olympics is not solid. The assuming of the relationships between CTs and air quality parameters in the same season (month) are constant in different years is not right. During the period 2000-2009 (except 2008), the background air pollutant concentration, the regional emission distribution and intensity have been changed significantly. Therefore, the relationships between CTs and air quality parameters in the same season will be changed. Why is SO2 so sensitive to synoptic circulation? According to the discussion at P33482, L14-15, SO2 concentration is more sensitive to emission rate rather than CTs.

Reply: We agree that there are large uncertainties of quantitative analysis of the impacts in circulation on air quality during the Olympics, which are originated in the emission variations, big variances of air pollutant concentrations in a specific CT. According to the referee's comment, we rectify the procedures for quantitative analyses in two ways: 1) the background data is based on the same period in summer from 2005-2009 (except 2008); 2) the air quality parameters are visibility, PM_{10} and BC because of the data duration and integrity. Since the gaseous species has shorter time duration, they may should be excluded for quantitative analyses. But as for a comparison to other air quality parameters, the evaluation of CT on gaseous pollutants during Olympics was temporarily reserved.

The important point of the quantitative analyses, calculating the percent that CT frequency anomaly during the Olympics could explain the reductions of air pollutants, is reserved. Although it has certain level uncertainty, the emission rate in the same season (summer) during 2005-2009 (except the emission control period) can be approximately assumed to be constant. The evaluation via chemical model still have large uncertainties that could originate from large bias of bottom-up emission inventory, the complexity of physical and chemical processes and subgrid-scale processes. Our newly developed method has some unique strengths; it can be easily used to objectively evaluate the meteorological effects on air quality based on established relationships between air quality and synoptic CT.

The variation of SO_2 emission during this period is significantly smaller than the variation caused by different circulation pattern. We suggest that the frequent very high concentration of SO_2 in winter could significantly elevate the averaged value in CT 1. The large seasonal variation of emissions could diminish the correlations between air quality and CTs.

18. P33486, L17-20. The two are not comparable.

Reply： Accepted. We delete it.

References:

An, X., Zhu, T., Wang, Z., Li, C., and Wang, Y.: A modeling analysis of a heavy air pollution episode occurred in Beijing, Atmos. Chem. Phys., 7, 3103-3114, 2007.

Chen, D. S., Cheng, S. Y., Liu, L., Chen, T., and Guo, X. R.: An integrated MM5–CMAQ modeling approach for assessing trans-boundary PM10 contribution to the host city of 2008 Olympic summer games—Beijing, China, Atmos. Environ., 41, 1237-1250, doi:10.1016/j.atmosenv.2006.09.045, 2007.

Cheng, Y., Heintzenberg, J., Wehner, B., Wu, Z., Su, H., Hu, M., and Mao, J.: Traffic restrictions in Beijing during the Sino-African Summit 2006: aerosol size distribution and visibility compared to long-term in situ observations, Atmos. Chem. Phys, 8, 7583-7594, 2008.

Chou, C. C. K., Tsai, C. Y., Chang, C. C., Lin, P. H., Liu, S. C., and Zhu, T.: Photochemical production of ozone in Beijing during the 2008 Olympic Games, Atmos. Chem. Phys., 11, 9825-9837, doi:10.5194/acp-11-9825-2011, 2011.

Huang, X. F., He, L. Y., Hu, M., Canagaratna, M. R., Sun, Y., Zhang, Q., Zhu, T., Xue, L., Zeng, L. W., Liu, X. G., Zhang, Y. H., Jayne, J. T., Ng, N. L., and Worsnop, D. R.: Highly time-resolved chemical characterization of atmospheric submicron particles during 2008 Beijing Olympic Games using an Aerodyne High-Resolution Aerosol Mass Spectrometer, Atmos. Chem. Phys., 10, 8933-8945, doi:10.5194/acp-10-8933-2010, 2010.

Huth, R.: An intercomparison of computer-assisted circulation classification methods, Int. J. Climatol., 16, 893-922, 1996.

Huth, R., Beck, C., Philipp, A., Demuzere, M., Ustrnul, Z., Cahynová, M., Kyselý, J., and Tveito, O. E.: Classifications of Atmospheric Circulation Patterns, Ann. N.Y. Acad. Sci., 1146, 105-152, 2008.

Philipp, A., Bartholy, J., Beck, C., Erpicum, M., Esteban, P., Fettweis, X., Huth, R., James, P., Jourdain, S., Kreienkamp, F., Krennert, T., Lykoudis, S., Michalides, S. C., Pianko-Kluczynska, K., Post, P., Álvarez, D. R., Schiemann, R., Spekat, A., and Tymvios, F. S.: Cost733cat – A database of weather and circulation type classifications, Physics and Chemistry of the Earth, Parts A/B/C, 35, 360-373, doi:10.1016/j.pce.2009.12.010, 2010.

Streets, D. G., Fu, J. S., Jang, C. J., Hao, J., He, K., Tang, X., Zhang, Y., Wang, Z., Li, Z., and Zhang, Q.: Air quality during the 2008 Beijing Olympic games, Atmos. Environ., 41, 480-492, 2007.

Wang, B., Shao, M., Lu, S. H., Yuan, B., Zhao, Y., Wang, M., Zhang, S. Q., and Wu, D.: Variation of ambient non-methane hydrocarbons in Beijing city in summer 2008, Atmos. Chem. Phys., 10, 5911-5923, 10.5194/acp-10-5911-2010, 2010.

Wang, M., Zhu, T., Zheng, J., Zhang, R., Zhang, S., Xie, X., Han, Y., and Li, Y.: Use of a mobile laboratory to evaluate changes in on-road air pollutants during the Beijing 2008 Summer Olympics, Atmos. Chem. Phys., 9, 8247-8263, 2009.

Wang, M., Zhu, T., Zhang, J. P., Zhang, Q. H., Lin, W. W., Li, Y., and Wang, Z. F.: Using a mobile laboratory to characterize the distribution and transport of sulfur dioxide in and around Beijing, Atmos. Chem. Phys., 11, 11631-11645, doi:10.5194/acp-11-11631-2011, 2011.

Wehner, B., Birmili, W., Ditas, F., Wu, Z., Hu, M., Liu, X., Mao, J., Sugimoto, N., and Wiedensohler, A.: Relationships between submicrometer particulate air pollution and air mass history in Beijing, China, 2004–2006, Atmos. Chem. Phys, 8, 6155-6168, 2008.

Wu, Q. Z., Wang, Z. F., Gbaguidi, A., Gao, C., Li, L. N., and Wang, W.: A numerical study of contributions to air pollution in Beijing during CAREBeijing-2006, Atmos. Chem. Phys., 11, 5997-6011, doi:10.5194/acp-11-5997-2011, 2011.

Xie，B., F. Zhang, Q. Zhang, J. Poterjoy and Y. Weng.: Observing Strategy and Observation Targeting for Tropical Cyclones using Ensemble-based Sensitivity Analysis and Data Assimilation. Journal of Geophysical Research, 2012, submitted.

Yan, P., Huang, J., and Draxler, R.: The long-term simulation of surface SO2 and evaluation of contributions from the different emission sources to Beijing city, Science in China Series D(Earth

Sciences), 48, 2005.

Zhang, Q., Streets, D. G., Carmichael, G. R., He, K., Huo, H., Kannari, A., Klimont, Z., Park, I., Reddy, S., and Fu, J.: Asian emissions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys., 9, 5131-5153, 2009.

Zhang, Q. H., Zhang, J. P., and Xue, H. W.: The challenge of improving visibility in Beijing, Atmos. Chem. Phys, 10, 7821-7827, 2010.

Zheng, J., Hu, M., Zhang, R., Yue, D., Wang, Z., Guo, S., Li, X., Bohn, B., Shao, M., He, L., Huang, X., Wiedensohler, A., and Zhu, T.: Measurements of gaseous H2SO4 by AP-ID-CIMS during CAREBeijing 2008 Campaign, Atmos. Chem. Phys., 11, 7755-7765, doi:10.5194/acp-11-7755-2011, 2011.

Zhu, C., Wang, B., and Qian, W.: Why do dust storms decrease in northern China concurrently with the recent global warming, Geophys. Res. Lett, 35, 2008.

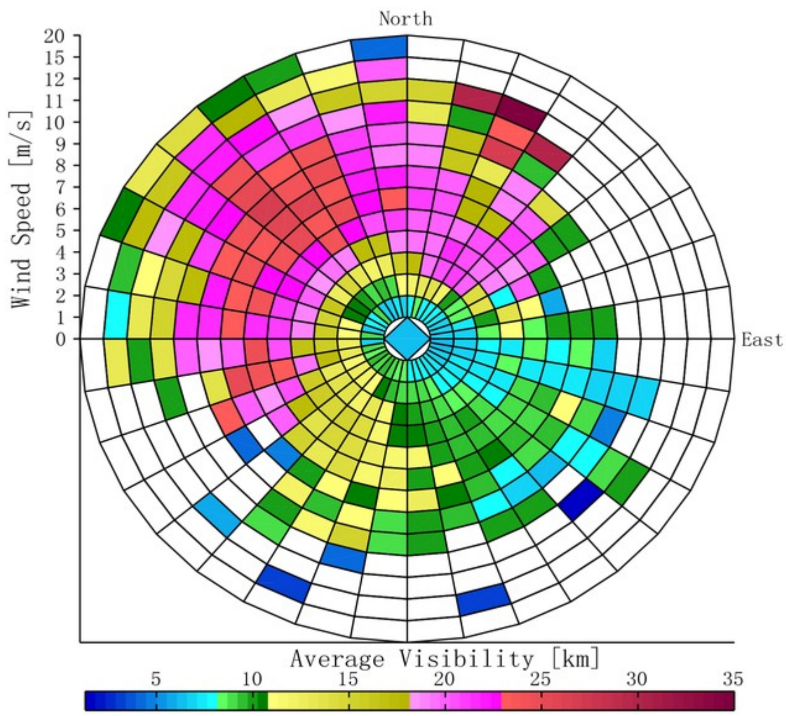

Fig. N7. Visibility distribution in relation to wind direction and wind speed at BCIA from 1999 to 2007. (from Q. H. Zhang et al., 2010, ACP)

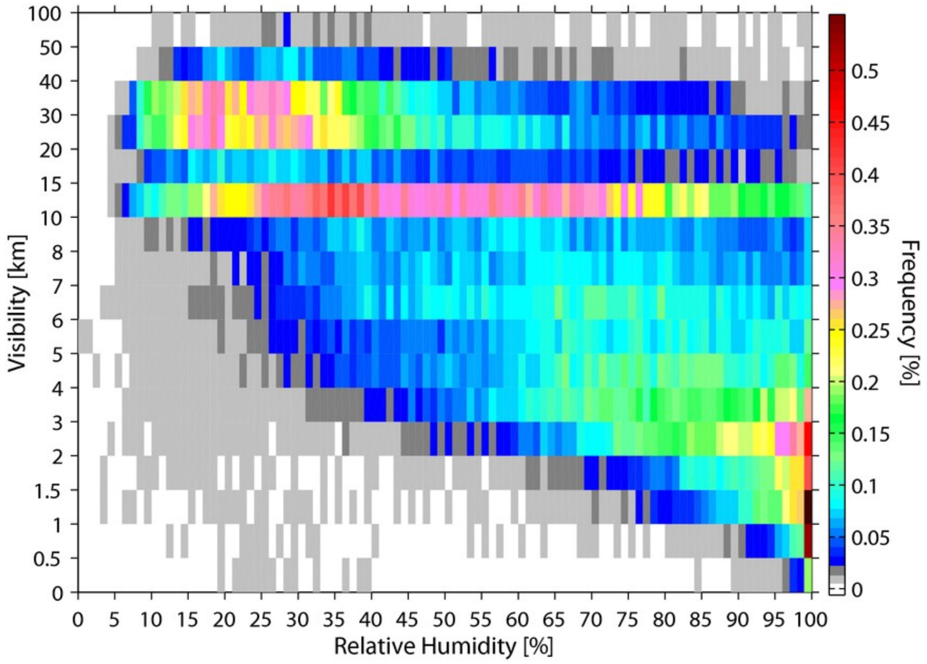

Fig. N6. Hourly occurrence frequency of visibility in relation to visibility and RH at BCIA from 1999 to 2007. (from Q. H. Zhang et al., 2010, ACP)

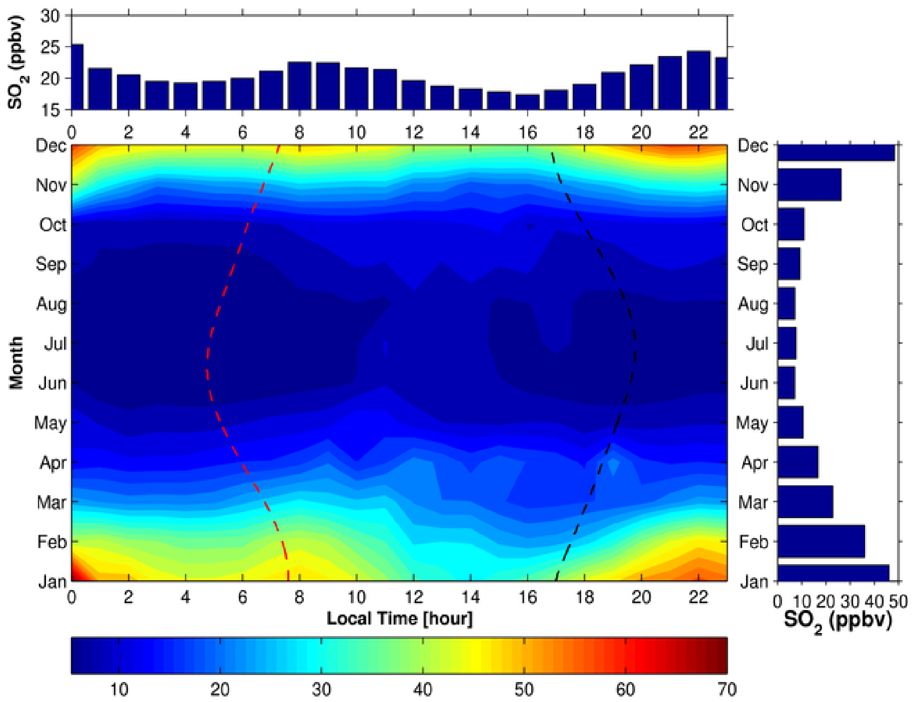

Fig. N8. SO_2 concentration distribution in relation to month and local time (filled color) from August 2006 to Octomber 2008. Red dash line represents sunrise time and black dash line represents sunset time.

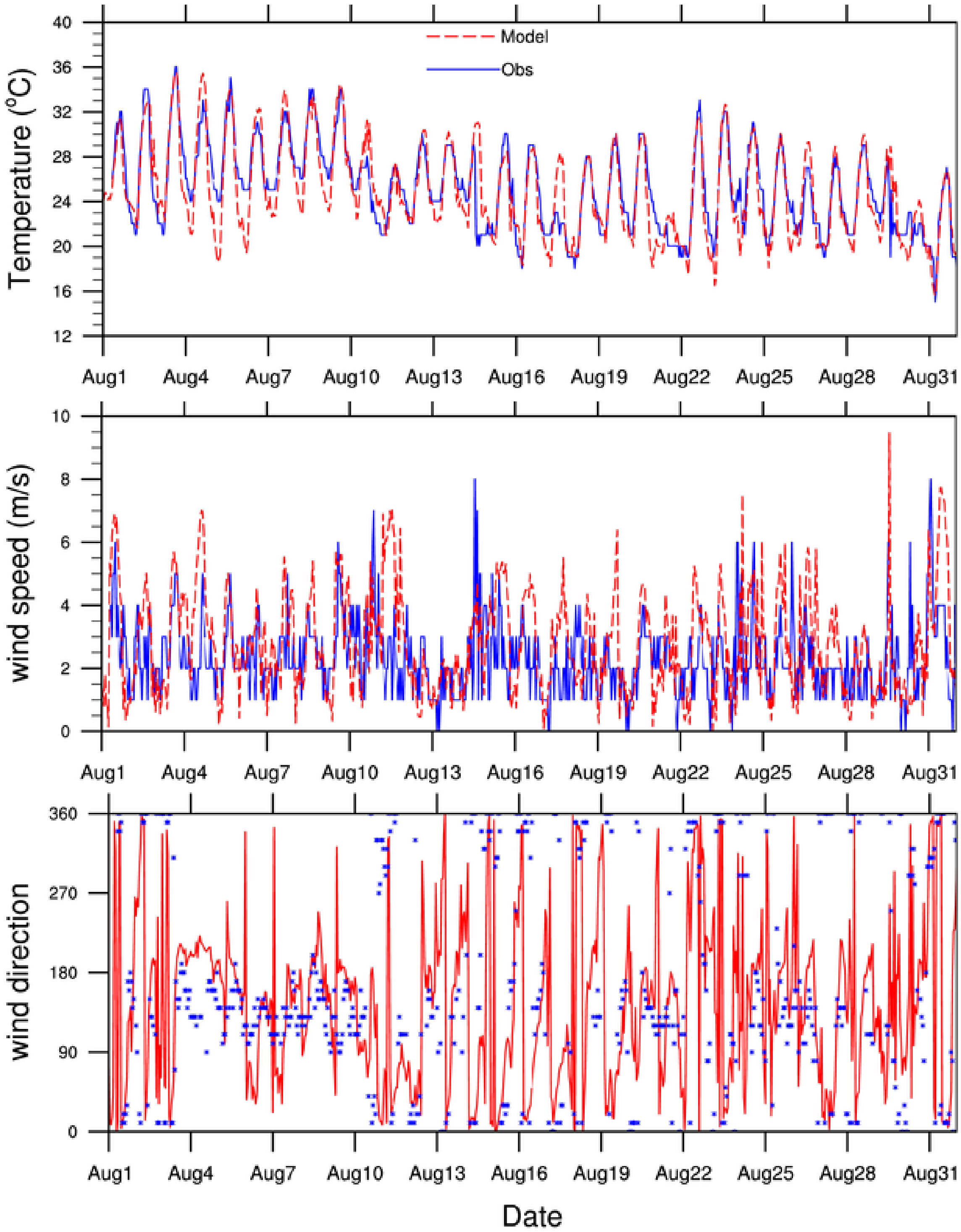

Figure S2(b). Comparison of model hourly outputs from 4 km WRF simulation (red line) with observed hourly temperature, relative humidity (at 2 m), wind speed and wind direction at 10 m (blue line or dot) at Beijing observation site (WMO 54511 site; 39.9°N, 116.3°E) for August 2008.

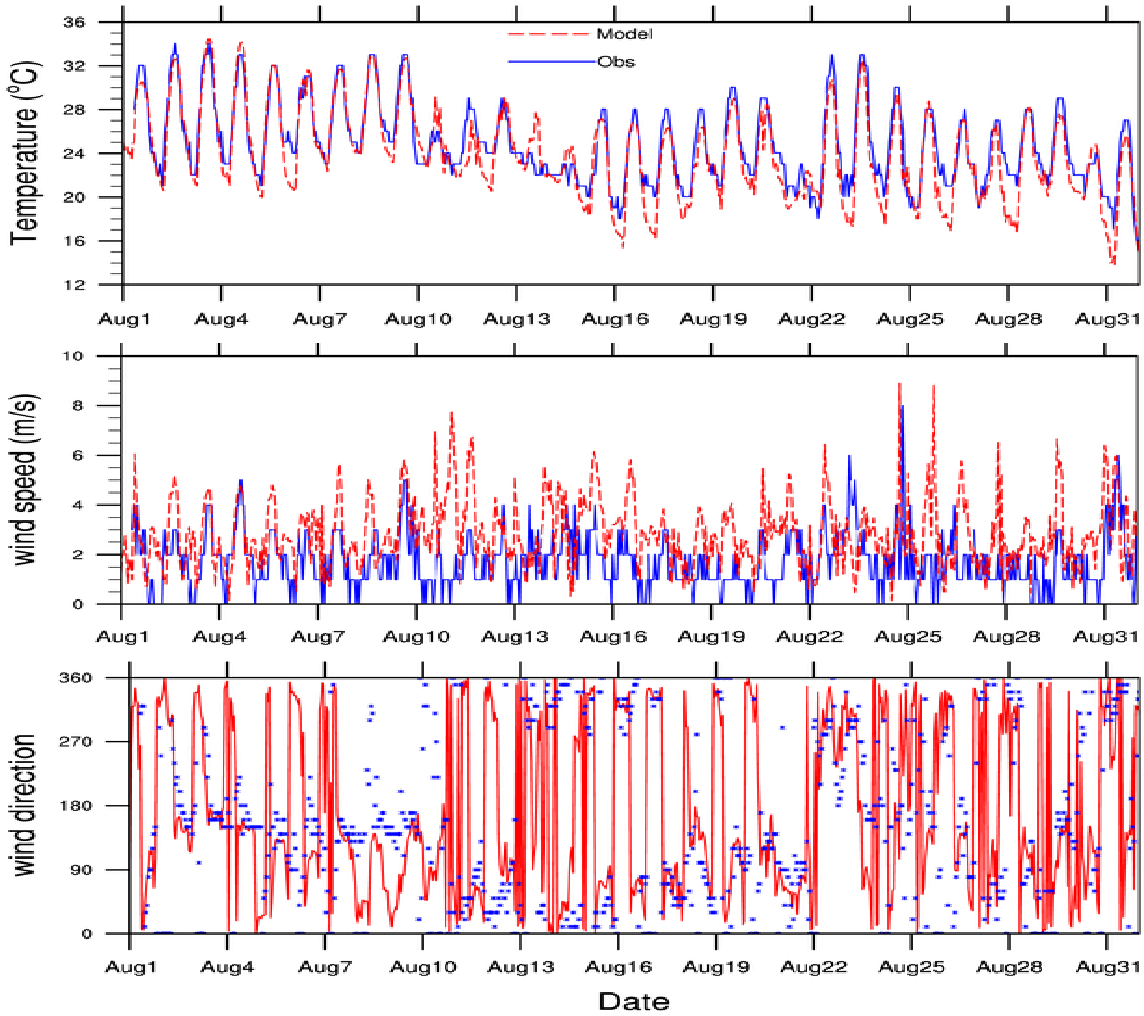

Figure S2(c). Comparison of model hourly outputs from 4 km WRF simulation (red line) with observed hourly temperature, relative humidity (at 2 m), wind speed and wind direction at 10 m (blue line or dot) at Shijiazhuang site (38.3°N, 114.7°E) for August 2008.

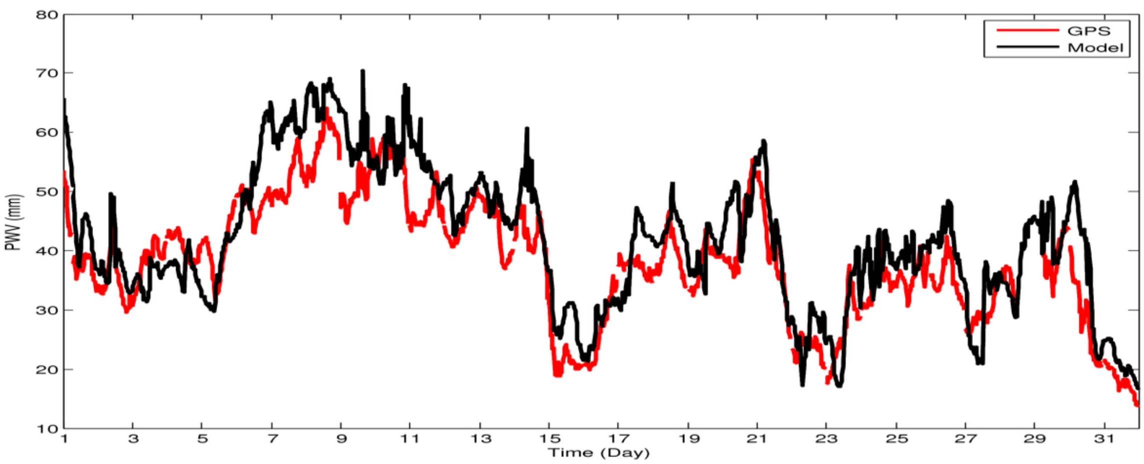

Fig. S2(d). Column-integrated Precipitable Water Vapor (PWV) at PKU site during August 2008. The GPS-based water vapor (data source: http://www.suominet.ucar.edu/support/) is shown in red line, and the black line denotes WRF-simulated water vapor (Domain 3). The correlation=0.884, mean error =3.62 mm.

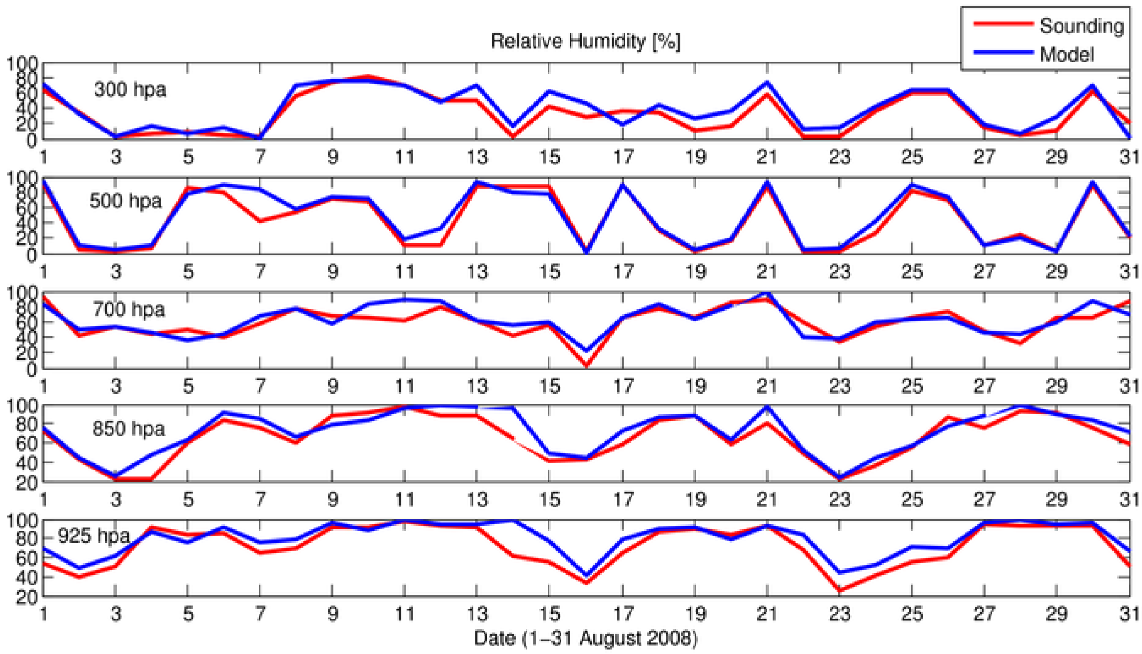

Fig. S21. Time series of observed (sounding-based measurements; red lines) and simulated (24h forecasting; blue lines) daily vertically relative humidity at Beijing Observational site. The bottom-up levels are 925, 850, 700, 500, and 300 hPa, respectively.

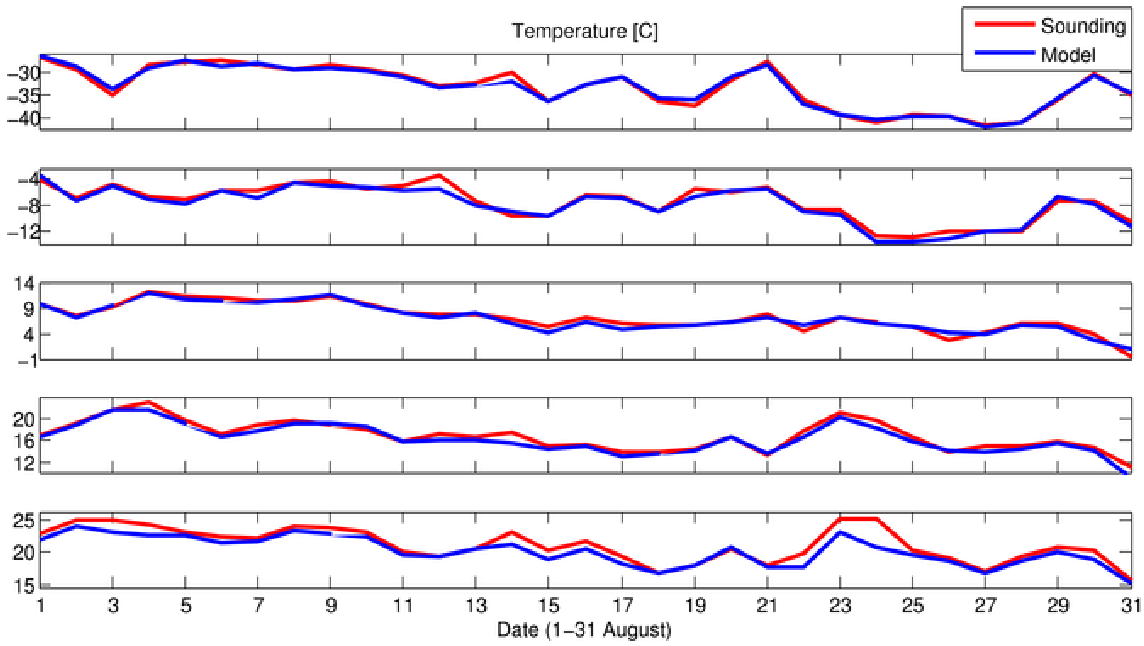

Fig. S22. The same as Fig. S21, but for temperature. The bottom-up levels are 925, 850, 700, 500, and 300 hPa, respectively.

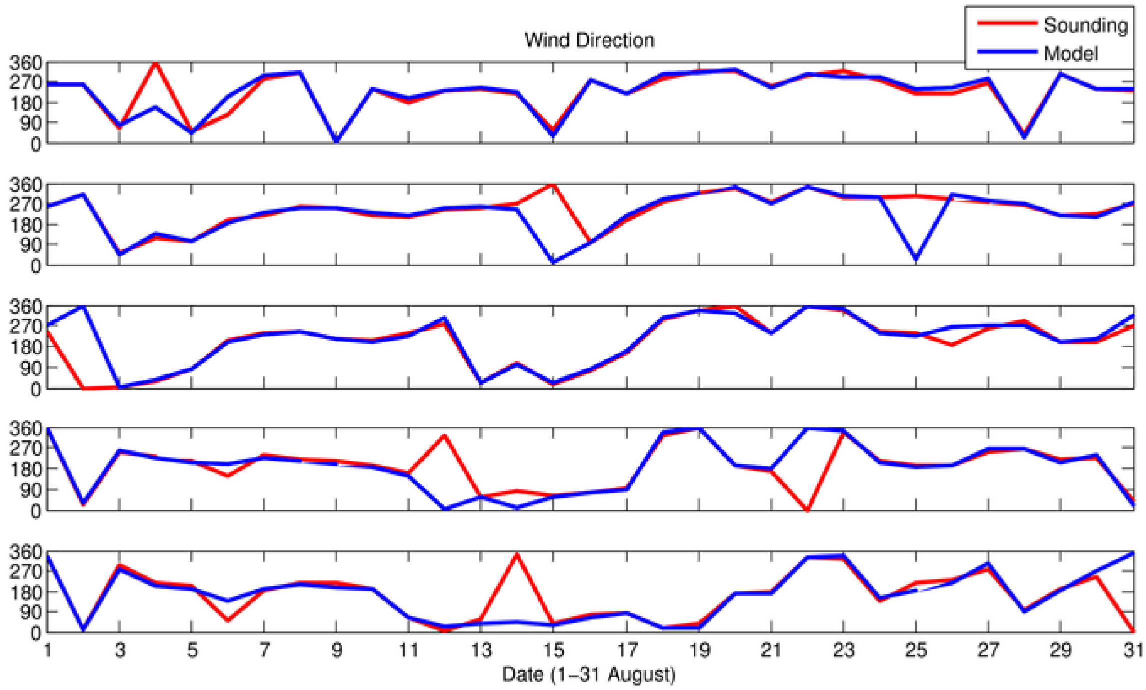

Fig. S23. The same as Fig. S21, but for wind direction.

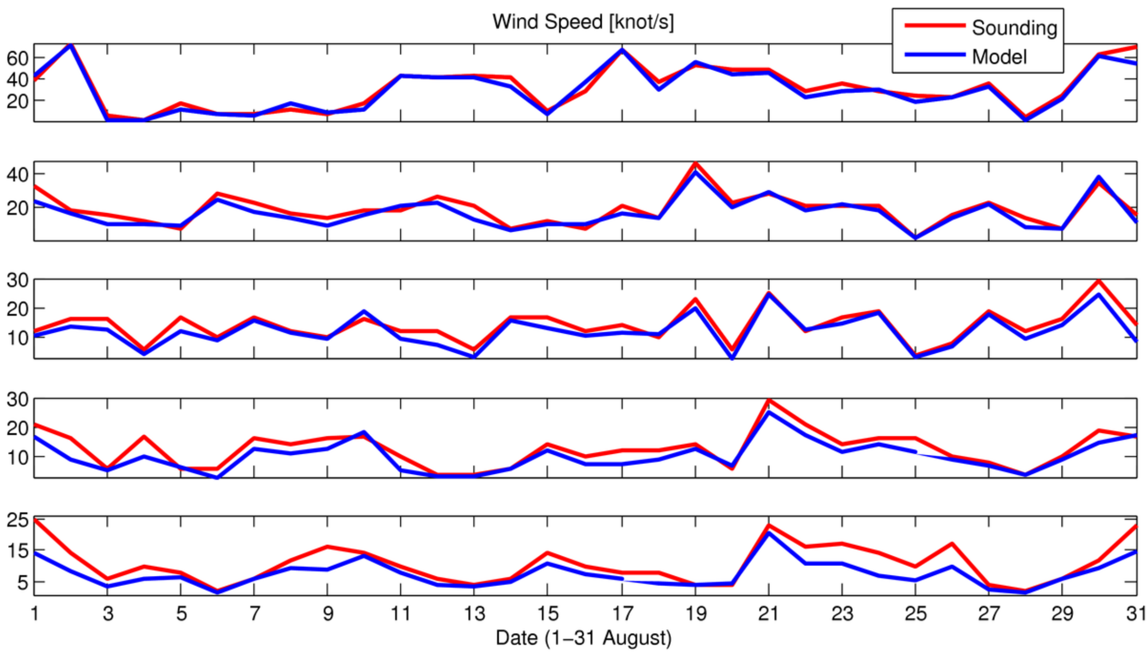

Fig. S24. The same as Fig. S21, but for wind speed.

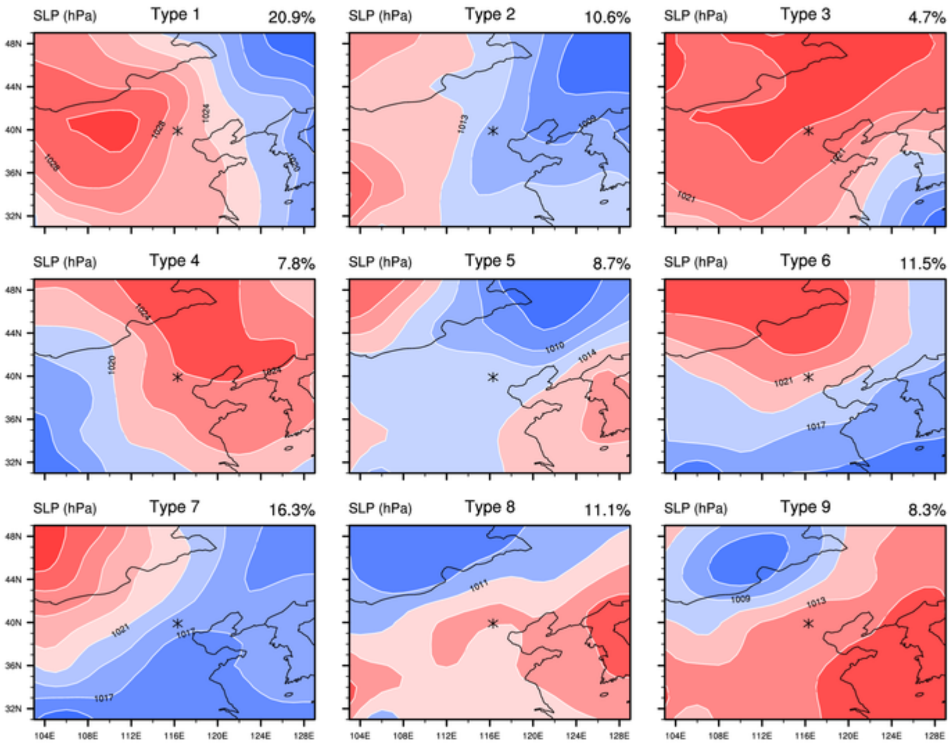

Fig. SN1. Mean sea level pressure (SLP) patterns and frequency of occurrence (right upper number) for each circulation type during the period 2000 to 2009. The asterisk represents the location of Beijing. Sensitivity study of adjustment of Domain 31-49N.

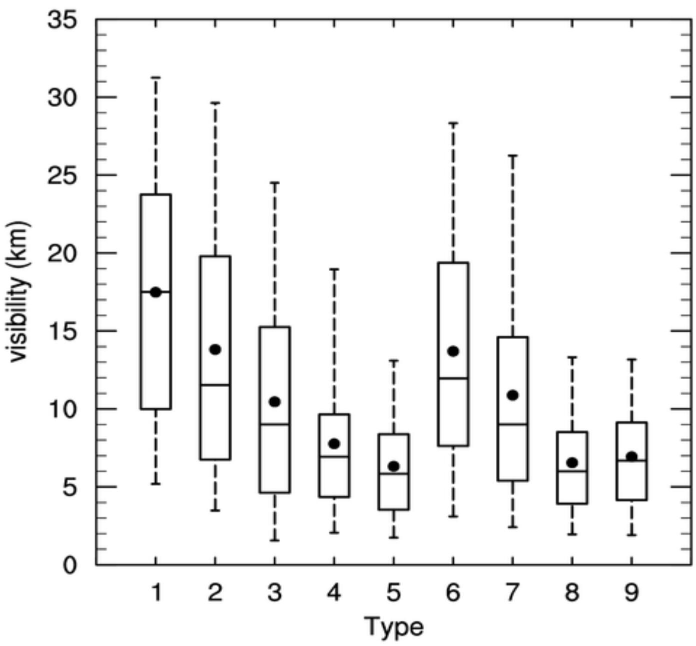

Fig SN2. Daily mean visibility at Beijing airport within the nine clusters. Sensitivity study of adjustment of Domain 31-49N.

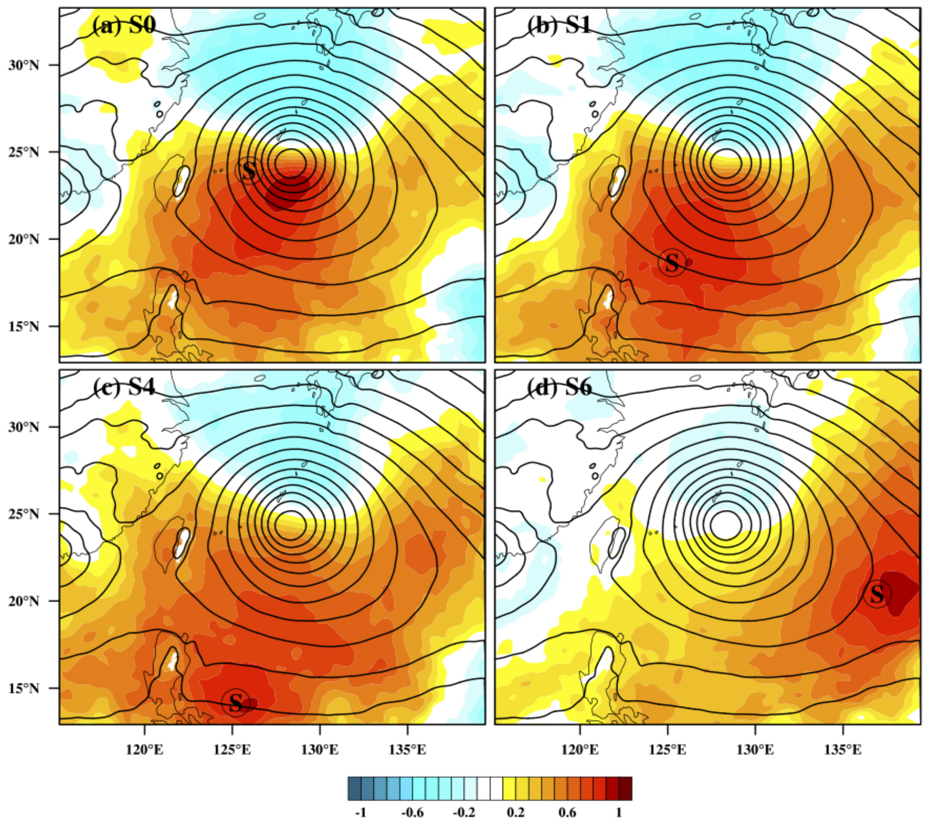

Fig. N5. The correlation between sea level pressure and 850 hPa height for 4 selected soundings. The position of sounding is marked as "Ⓢ". (from Xie et al., 2012, submitted to JGR)

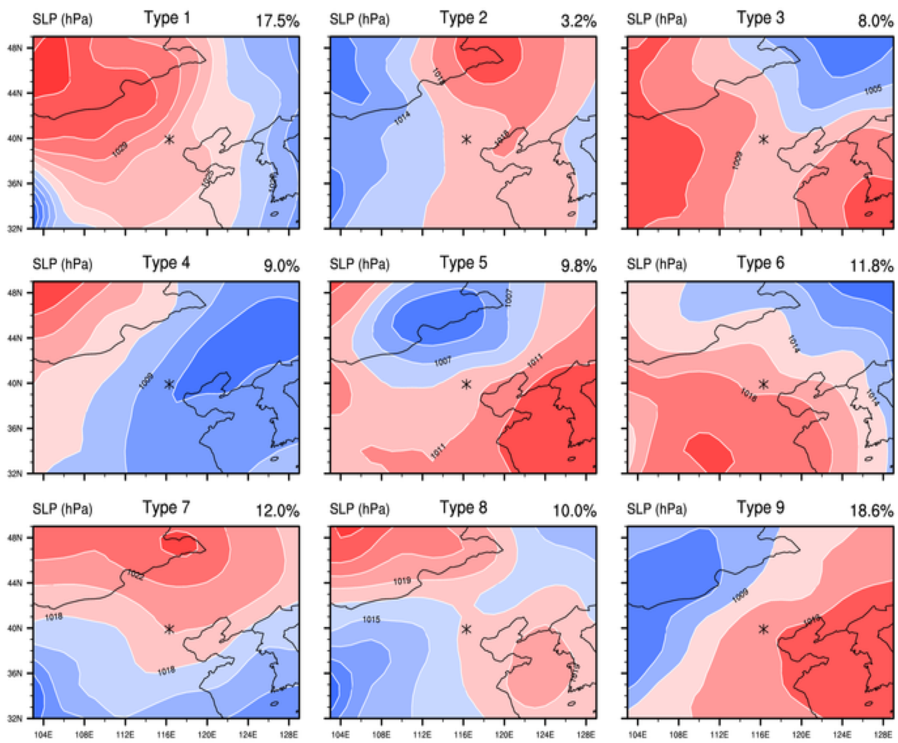

Fig. SN3. Mean sea level pressure (SLP) patterns and frequency of occurrence (right upper number) for each circulation type during the period 2000 to 2009. Sensitivity study of adjustment input time as 06:00 UTC (LT 14:00).

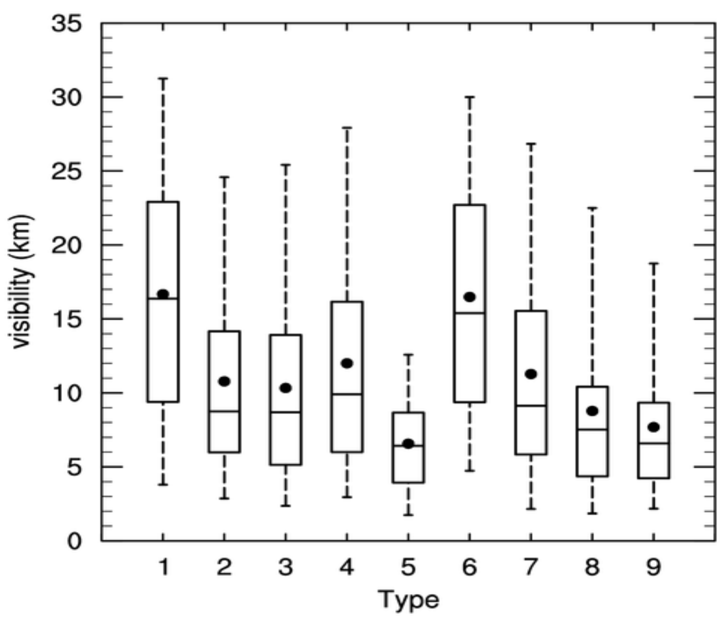

Fig SN4. Daily mean visibility at Beijing airport within the nine clusters. Sensitivity study of adjustment input time as 06:00 UTC (LT 14:00).