Atmos. Chem. Phys. Discuss., 11, C15296–C15303, 2012
www.atmos-chem-phys-discuss.net/11/C15296/2012/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "The impact of circulation patterns on regional transport pathways and air quality over Beijing and its surroundings" *by* J. P. Zhang et al.

Anonymous Referee #2

Received and published: 14 February 2012

General Comments:

In this work, the authors investigate how different regional circulation patterns are correlated with different environmental variables over Beijing. The authors used principal component analysis, in connection with reanalysis data of surface pressure variables over East Asia, to look at orthogonal air circulation patterns. They then use this daily average circulation classification to look at how it is correlated with different measurements of some visibility and pollution records.

This is a very interesting and important work, one that has a good potential to help guide further science and policy developments. I would ultimately like to see the results

of this effort published. However, given the current issues that I have with many of the details of how these correlations were established, and given that many issues exist with the fundamental statistics and treatment of the modeling environment, at the present time I recommend that the paper be rejected. I would strongly encourage the authors to re-work through the following issues, and if successful, to resubmit their work again.

One specific reason for rejection is that many of the correlations are based on inappropriate statistics, and therefore I believe may not be valid. For example, the authors are attempting to correlate the daily average meteorological fields with AOD from AERONET, yet the AERONET daily average data from the stations mentioned is missing from more than half of the days. While it may be possible to look at the correlations on a month by month average, this is not necessarily easy to do, due to gaps in the data. A similar argument is made for comparison with the MODIS data, which has a far lower frequency than that of AERONET over this environment.

A second specific reason for rejection is that nowhere in this effort are considerations for important effects other than transport and meteorology considered. There seems to be no consideration for the state of the boundary conditions, nor does there seem to be consideration of the highly non-linear chemistry and physics that the aerosols and reactive gasses undergo. While some of these are based at least in part upon the temperature, relative humidity, and rates of mixing, others need to be resolved at the grid scale. There have been many efforts to look at this, both in a Lagrangian as well as Eulerian framework, and this must be taken into account, if we are to try to accomplish any type of correlation between the impacts of meteorology and the observed results.

Specific Comments:

It seems that reanalysis data (at 1ox1o) has been used for downscaling with WRF to higher resolution. I am wondering why it is that the model has not been run using a more typical approach whereby the regional model is run continuously over the entire

ACPD
11, C15296–C15303, 2012

period from 2000-2009, with updates at the boundaries and some type of nudging applied. I have not previously seen the model run with continuously updating initial conditions, just for 1.5 days, and then reset each day. Perhaps there is precedent for this, but if it is the case, I would like to see this documented. I would be interested to know, in this case, if there are gaps or other differences in the fields observed from day to day?

The authors state that PM10 and BC are measured from the time period from 2005-2009. Yet their correlations are used against meteorological data from the time period from 2000-2009. Are the results the same when the same time periods are considered? Similarly, with respect to O3, SO2, NO2, and CO measurements, which seem to have been from gathered only from 08/2006-10/2008.

The 500nm AOD data from the AERONET sites Beijing and XH have many individual days missing from their data, when looked at over the period from 03/2001-10/2008. How are these missing data points considered? It seems that the coverage during certain months of this spanning data set are quite small, and in some cases do not exist. Does this allow for a statistically representative correlation with the daily circulation type to be accomplished?

What is the top of the domain used in the WRF downscaling at the 36km, 12km, and 4km grids? Is it sufficiently high to capture the effects of the transport of Dust from the Northwest, which are known to make an important contribution to the aerosol loading over Beijing.

Some of the comparisons between the WRF fields do not seem to compare well against some of the meteorological variables from the airport. Does this disagreement potentially translate into the EOF decomposition for the circulation types?

There are three important issues built into the use of FLEXPART that make it problematic in this case, and extremely important to address if it is used in this context. (1) Given that the lifetime of aerosols and some of the gas-phase tracers looked at here

ACPD

11, C15296–C15303, 2012

are a few days to a week, why has FLEXPART only been run for 2 days? (2) Since some of the pieces of data being compared against are column values, such as AOD, should not FLEXPART be run backwards throughout the atmosphere, or at least up to the middle troposphere. Given that dust is an important source of both PM10 and AOD over Beijing, and given that most is imported by long-range transport, it would seem important to consider this contribution. (3) FLEXPART is a fair model for following relatively unreactive species, such as air molecules and CO, however, when being used for highly reactive species, diffusion to and from surrounding parcels, as well as removal from or addition to the parcel due to wet uptake, evaporation, condensation, coagulation, etc. need to be accounted for. How is that done in this case, and if it is not done, how much error would it add?

Why are back trajectories limited to the 0-50m levels? Is there some reason to believe that the local boundary layer is not better mixed than that, with respect to a daily average measurement value (the same scale that is being correlated against)? Is there an issue with local sources being overly sampled at the site that requires only the very bottom of the atmosphere to be considered as being the sample range at the site?

Atmospheric visibility is a combination of aerosol concentration, aerosol size and chemical composition, relative humidity, temperature, and other factors. Since the comparisons are being made with strictly the meteorological variables, how relevant are each of these terms? Which meteorological term is more important or the dominating contribution for each correlation?

Higher PBL will lead to a change in the surface concentrations, if the boundary layer is well mixed. However, if this is the case, then as mentioned above, the FLEXPART runs should be done at least throughout the entire boundary layer for concentration measurements. Further, the effect of the boundary layer height on column values, such as AOD, is not so straightforward.

Your comments on AOD do not match with all of the observations. It is my understand-

ACPD

11, C15296–C15303, 2012

ing that at least some of the highest AOD events in Beijing have been when wind blows large amounts of desert dust, which is most similar to CT1. Also, very stagnant or re-circulating conditions have led to very high AOD levels. However, since this is an effect of long-range transport in the first example, and longer-lifetime aerosol being recycled in the second example, how can these be incorporated into your modeling simulation?

AOD is a column value, and is strongly influenced by aerosols throughout the entire column, not just near the surface.

MODIS AOD at 1km is a model product. Since there are not too many cloud-free or uncontaminated clouds, it has a very low reliability in terms of AOD. Furthermore, even where it exists, it tends to have quite poor statistics even on a monthly average value. It would not be considered trustworthy on a day-to-day type of basis at all.

Although your mean PM10 values look different, their error bounds seem wide. Are the differences statistically significant?

BC is more closely correlated with dispersion, but under heavily polluted urban regions, the chemistry is still quite important. This is all the more important since chemical and physical ageing of BC alter its lifetime and atmospheric properties, and cannot be determined from local conditions alone (eg: Kim et al 2008; Cohen et al. 2011). It seems quite interesting that CT1 has a relatively high amount of SO2, although overall it tends to have lower amounts of other species. Is there a difference in the correlation found between the high average SO2 days and BC in CT1, as compared to the low SO2 days and BC in CT1?

Why is the SO2 so much higher, as compared with most of the other species, in the clean case CT1, unless either the emissions of SO2 are higher, or unless the chemical destruction of SO2 is lower? If the emissions are assumed to be constant, then the chemistry must be important. This is an important point that needs to be addressed.

High levels of sulfate indicate that the air has had a somewhat long residence time

ACPD

11, C15296–C15303, 2012

in the atmosphere, since it has to have time to chemically form. High levels of SO2 indicate a short residence time in the atmosphere, or special conditions under which the SO2 has not been able to oxidize to form sulfate. How do you resolve the fact that some of the CTs seem to have considerably different ratios of sulfate to SO2, unless either or both of long-range transport and chemistry are considered?

How much of this correlation between the southerly airflow into Beijing is related to a net mass flux of pollutants into Beijing, as compared with the fact that the warmer air will cause secondary aerosol chemistry to go faster?

The Beijing site AERONET data is not sufficient, on a daily resolution, to make such a broad statement. The total number of individual daily measurements made at this site, from 2002 through 2010, during August are not sufficient to cover even a reasonable fraction of the daily average AOD values. How such a strong conclusion can be drawn seems unlikely. The conclusions of the daily variability cannot be supported using this set of data for this case.

Since policies enacted lead to a different condition in 2008, looking at the average of non-2008 data and then making a comparison with the 2008 data could be justified. However, in terms of the meteorology, this is not true, unless people had an impact on the large-scale circulation. Therefore, it is not statistically correct to look at the anomaly of the meteorology excluding 2008 as compared with 2008, and instead the value in 2008 should be compared with the baseline value over the entire time span being looked at.

Reductions or increases in NO2 are much more difficult to understand as compared with NOx or NOy. Emissions reductions could lead to an increase or a decrease on NO2, based on the various different changes in other gas-phase chemicals, and therefore, are not very useful when making such comparisons.

You mention that your result is for visibility is 50%. Yet you mention that Q. H. Zhang et al. 2010 find that RH contributed 24%. However, visibility is a combination of both

ACPD

11, C15296–C15303, 2012

changes in aerosols as well as changes in water uptake. If they found, as you state, that there was a lower amount of hydroscopicity, then this would imply a higher ratio of BC and other hydrophobic aerosols to hydrophilic aerosols. This is a very different conclusion: it means that the aerosol concentrations were not kept in the same proportion. I know that you have mentioned sulfate in the paper, but I do not see a careful examination of how it has correlated with the different CTs. I think that this is an important point that must be addressed, if the conclusion is to be valid whether it is circulation, aerosol physics, or hydroscopic effects. Just stating that it is all due to meteorology is an oversimplification.

Figure 7: How many different days are being compiled? What is the fraction of the total days?

Figure 9 shows that the concentrations of PM10 and BC are generally lowest in CT1 and CT6, as claimed in the paper, although with a large amount of variability. Yet, in Figure 10, it shows that for CO, the best of the chemicals present in this report to use as a "pure tracer" (due to its relatively longer chemical lifetime) that the lowest concentrations are in CT6 whereas CT1 seems quit similar to many of the other types. SO2 seems to be relatively higher in CT1 than the other types. These conclusions lead me to believe that chemistry and aerosol physics are important processes occurring here. On the other hand, it could also be an artifact of how gaps in the data were dealt with. Are these values presented, as mentioned here, daily averages, and if so, how were they computed? Are there specific times of day or days of the year that were not systematically measured less frequently?

Technical Corrections:

33779(6-7): Mixing of air does not reduce the amount of aerosols, what it does is increases aerosols in some places while reducing them in others, smoothing out gradients in concentration. An increase in wind speed also does not reduce the total amount of aerosols, it merely causes them to be transported elsewhere.

ACPD

11, C15296–C15303, 2012

33480(3): AOD values of 0.43 are not that low. These are already somewhat polluted condition, where the chemistry and physics of the aerosols present cannot be neglected.

33483(3): Why during the Olympic period are only 24 hour back-trajectories used, as compared to the 48 hour back-trajectories used during the other times?

References:

Cohen, J. B., R. G. Prinn, and C. Wang, 2011: The impact of detailed urban-scale processing on the composition, distribution, and radiative forcing of anthropogenic aerosols, Geophys. Res. Lett., 38, L20808, doi:10.1029/2011GL047417.

Kim, D., C. Wang, A.M.L. Ekman, M. C. Barth, and P. Rasch, 2008: Distribution and direct radiative forcing of carbonaceous and sulfate aerosols in an interactive size-resolving aerosol-climate model, J. Geophys. Res., 113, D16309, doi:10.1029/2007JD009756.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 33465, 2011.