Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-911-AC1, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Detecting critical $PM_{2.5}$ emission sources and their contributions to a heavy haze episode in Beijing, China by using an adjoint model" *by* Shixian Zhai et al.

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Received and published: 14 February 2017

Dear referee,

Thank you very much for your valuable comments. Revisions made in the manuscript are in red characters or highlighted. Responses to the comments are listed as followings:

General comments

The authors attempted to determine the contributions from local and surrounding emission to two PM2.5 peaks during a heavy Beijing haze episode by using an aerosol adjoint model. Sensitivity analysis of the model simulations was performed to detect the

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Discussion paper

PM concentration-source relationship by examining the temporal variation of a user-defined sensitivity coefficient and its time-integrated values. Given that there are still debates on the relative contributions of aerosols from local emission and regional transport to Beijing haze, the adjoint modeling studies and sensitivity analysis in this study would be interesting to the readerships of the ACP journal. However, some issues related to the clarity of discussions need to be addressed before its publication.

Specific comments

1) According to Fig. 4, what the reasons for the significant decrease of PM concentration during 11:00 to 17:00 on Nov. 21st? Could it be the development of PBL or the reduction of emissions during this period?

Response: Decrease of PM2.5 concentration from morning to afternoon is typical in Beijing, resulted mainly from diurnal variation of (planetary boundary layer) PBL with the development of vertical mixing after sunrise for diluting pollutants (Zhao et al., 2009; Liu et al., 2015). Meanwhile, the emissions could be reduced during this period (An et al., 2013).

P6: lines 20-26 in section 3.4 were revised as: Thereafter, PM2.5 concentration over Beijing, south central Hebei and Tianjin decreases to a trough in the Nov. 21st afternoon before rising above 550μg/m3 at 23:00 BT. Decrease of PM2.5 concentration from morning to afternoon is typical in Beijing, resulted mainly from diurnal variation of (planetary boundary layer) PBL with the development of vertical mixing after sunrise for diluting pollutants (Zhao et al., 2009; Liu et al., 2015). Meanwhile, the emissions could be reduced during this period (An et al., 2013). The concentration peak at 23:00 BT owns to the influence of the easterly winds on Nov. 21st, which causes pollutants convergence ahead of the Tai-hang Mountains and carries abundant water vapor that promotes local hygroscopic growth.

References:

An, X. Q., Sun, Z. B., Lin, W. L., Jin, M., and Li, N.: Emission inventory evaluation using observations of regional atmospheric background stations of China, J. Environ. Sci., 25, 537-546, 2013.

Added references:

Liu, Z., Hu, B., Wang, L., Wu, F., Gao, W., and Wang, Y.: Seasonal and diurnal variation in particulate matter (PM10 and PM2.5) at an urban site of Beijing: analyses from a 9-year study, Environmental Science and Pollution Research, 22, 627-642, 2015.

Zhao, X., Zhang, X., Xu, X., Xu, J., Meng, W., and Pu, W.: Seasonal and diurnal variations of ambient PM2.5 concentration in urban and rural environments in Beijing, Atmospheric Environment, 43, 2893-2900, 2009.

2) On p. 10 lines 2-5, the authors attributed the overall higher contribution from the surrounding emissions than local emissions to the obvious periodic fluctuation of hourly sensitivity coefficient of surrounding emissions. This explanation is not convincible for me since we only can infer that there was larger temporal variation for the contribution of surrounding emissions than that of local emissions based on the fluctuation of sensitivity coefficient.

Response: Thank you for the comments. Here we are aiming to find out the more influential regions and time period of emissions from the hourly sensitivity coefficients. Since we can infer 'that there is larger temporal variation for the contribution of surrounding emissions than that of local emissions based on the fluctuation of sensitivity coefficient', surrounding emissions emitted 2-3 days ahead of the pollution peak is of great importance to the pollution peak . In this way, joint control of emissions with surrounding areas ahead of the polluted peak can lead to effective result.

To clarify this statement, we have revised these lines to: From the time series of hourly sensitivity coefficients for local and surrounding emissions (Fig. 6a and b), we can see that the temporal variation in the contribution of surrounding emissions is larger than

that of local emissions during 2-3 days ahead of the pollution peak, which indicates a notable contribution of surrounding emissions to PM2.5 peak during this time period. Therefore, if joint control of pollutant emissions with Hebei province, Tianjin city and Shanxi province were implemented 2-3 days ahead of the first PM2.5 concentration peak, then PM2.5 concentration accumulation due to transported pollutants could be effectively prevented, thus decreasing the concentration of these two PM2.5 concentration peaks.

3) I would suggest the authors to move the detailed discussions about computational efficiency of the adjoint model and the Models-3/CMAQ systems in the conclusion section (p. 12 lines 3-9) to section 4.3. Just a brief and concise summary is needed for the model computational efficiency in the conclusion section.

Response: Thank you for your valuable suggestion, this will definitely make section 4.3 more complete and the conclusion part concise.

We have moved the following content as an added paragraph at the end of section 4.3: Beyond that, the computational loads of the adjoint simulation are much smaller than the assessments with Models-3/CMAQ modeling (Zhai et al., 2016). For the adjoint simulation, one forward integration (for un-equilibrated data saving) and one backward adjoint integration can obtain the influence of emissions from any source region, during any time period to PM2.5 peaks. However, in the Models-3/CMAQ assessments, in order to compare the effects of emission reductions over two different time periods, at two different ratios and over four different regions, 12 sensitivity tests are set and the forward model is integrated for 13 times (one control simulation included).

We also replaced the above contents in the conclusion with a concise sentence: Meanwhile, the adjoint simulation is of much higher computational efficiency than the assessments with Models-3/CMAQ modeling.

4) It was stated that the threshold to determine sensitive emission regions was based on the relative magnitude of sensitive coefficients and the sources contribution ratios

of sensitive regions to the objective function (p. 10 lines 21-23). What are the exact values for the relative magnitude of sensitive coefficients and the sources contribution ratios of sensitive regions to the objective function? Otherwise, I feel that the selection of the threshold is arbitrary.

Response: Thank you very much for pointing this out. Here, we want to express the threshold in order to compare the adjoint results and the Models-3/CMAQ assessments (Zhai et al., 2016), the full administrative regions (HuaB and BJ) and the sensitive regions (HuaB-sens and BJ-sens) are selected based on the regions definition in the previous research (Zhai et al., 2016) and the adjoint results. The reason why we choose 3μg/m3 as the threshold is that in this way, the HuaB-sens accounts for 10.2% the area of HuaB and the BJ-sens accounts for 60.0% the area of BJ, similar with the sensitive and administrative regions definition by Zhai et al. (2016), thus making the results comparable.

We have revised the the corresponding description to: In order to compare the adjoint results with the Models-3/CMAQ assessments, we refer to the research by Zhai et al. (2016) and select four emission regions based on administrative divisions and the adjoint results: the overall Huabei region (HuaB), the sensitive Huabei region (HuaB-sens), the overall Beijing municipality (BJ), and the sensitive Beijing region (BJ-sens) (Fig. 8). In this research, grid cells with 72-h cumulated sensitivity coefficient larger than 3μg/m3 are covered by the sensitive emission regions (HuaB-sens and BJ-sens), and grid cells with smaller sensitive values are outside the sensitive emission regions. This means that emissions within the sensitive emission regions have relatively larger impact on the PM2.5 concentration peak. Here, the HuaB-sens accounts for 10.2% the area of HuaB and the BJ-sens accounts for 60.0% the area of BJ (Fig. 8), similar with the sensitive and administrative regions definition in research by Zhai et al. (2016).

Technical corrections

1) On p. 11 line 19: remove the first ‘peak’.

Response: The ‘peak’ is removed.

2) The text in Fig. 1 was not legible. Please enlarge the font size.

Response: The brief explanations of Fig. 1 were added in the Fig. 1 caption as follows: Figure 1. (a-d): Sea-level pressure field (black contour lines; Beijing is marked with a red triangle); (e-h): temperature-logarithmic pressure diagrams (thick red solid curves for: process; green solid curves for: dew point-pressure; blue solid curve for: stratification) at Nanjiao Station from 08:00 (local time) on Nov. 20th 2012 to 20:00 (local time) on Nov. 21st 2012. Detailed information of Fig. 1 is found in Fig. S1 in the supplement. Meanwhile, enlarged figures and explanations were added in Fig. S1 in the supplement. Correspondingly, ‘Fig. S1’ is revised to ‘Fig. S2’, and ‘Fig. S2’ is revised to ‘Fig. S3’ in the manuscript.

3) For Fig. 6, the Y axis label of “PM2.5 concentration” overlays on the one for panel (c). Adjust its position to where is only for panels (a) and (b).

Response: The position of the Y axis label of “PM2.5 concentration” was adjusted.

4) P. 6 line 8: ‘might attribute’ should be ‘might be attributed’.

Response: ‘might attribute’ was changed to ‘might be attributed’.

5) P. 6 line 10: I would like to use ‘have proven’ to replace ‘had convinced’.

Response: Thank you for your suggestion. The ‘had convinced’ was replaced by ‘have proven’.

6) Please pay attention to the tense consistency through the manuscript. For example, on p. 6 line 28, it used both current and pass tenses (note the words ‘take’ and ‘defined’).

Response: We have revised the whole manuscript to current tense. Revised words are in red color.

Other minor revisions

P2 line 6: ‘to any model parameters’ is changed to ‘to model parameters’.

P3 line 27: ‘the ahead 2 days’ is changed to ‘the previous 2 days’.

P4 line 27: ‘reflect exact contribution’ is revised to ‘reflect the absolute contribution’.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-911/acp-2016-911-AC1-supplement.zip

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