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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.

Anonymous Referee #3

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1 Overview

The manuscript by Zhai et al. investigates sources of $PM_{2.5}$ for a pollution episode in Beijing using adjoint modeling. The work is a nice start, and a good use of the new tools that this group has developed. However, the manuscript overall feels a bit premature; it reads like a first draft. The overall purpose of using the adjoint model is not well articulated, nor is the tool used to its full potential. The comparisons and evaluations to observations and other studies are often qualitative and not particularly well fleshed out, and the presentation of results is murky in a few critical areas. The manuscript also requires substantial grammatical editing throughout. It is possible the

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work would be suitable for ACP after major revisions, but a different journal such as Atmospheric Research may be a better fit.

2 Major comments

- 2.10-14: While it is true that an adjoint model provides more precise estimates of the sensitivity (partial derivative), this in some cases may also be viewed as a downside compared to perturbation approaches when performing sensitivity calculations for the purpose of source attribution, since the adjoint model fails to capture the nonlinear response of atmospheric chemistry to substantial changes in emissions. Overall, the topic of how these types of sensitivities are interpreted for source contributions needs to be directly addressed in the introduction and methods, and expanded upon in the interpretation of results in more detail.

- Introduction: Several previous studies of source contributions to $PM_{2.5}$ in Beijing are mentioned, but they are only discussed in terms of their computational methods. That would be fine if this paper was in G.M.D. and strictly a discussion of methods. But for a scientific paper in ACP, the authors need to discuss the actual scientific findings of previous works. They need to clearly articulate what has previously been written about the sources that contribute to Beijing $PM_{2.5}$, and how their current study will advance the understanding of sources (most likely by providing insights into the spatial variability of contributions that can be most readily obtained using adjoint methods). Some justification for studying the specific pollution episode of Nov 19-21, 2012, also needs to be provided.

- 5.26: The authors claim that the initial concentrations and boundary conditions are set as the “observed monthly means”, but this does not make sense, as it is impossible that the concentrations of all species were observed at all locations throughout the domain in order to established an observationally-derived initial

condition and boundary condition. Thus, please describe in more detail how initial and boundary conditions are estimated.

- Section 3: Please include an entire new section covering in detail the emissions (anthropogenic and natural) used in this model, including a description of their daily and hourly variability. These are critical for understanding the significance of sensitivities of the form $\frac{\partial J}{\partial S_n} S_n$.

- Fig 3(a) / Section 3.3: The discussion model performance evaluation needs to be improved and expanded. It appears that the simulation over-estimates $PM_{2.5}$ concentrations are overestimated, although the timing of the peaks is well-correlated with the measurements. Are there no measurements on Beijing site to compare with? Are only measurements of total $PM_{2.5}$ available? How well does this model do at reproducing concentrations of specific aerosol components, such as BC, sulfate, nitrate, etc.? If this has been documented in previous work for Beijing specifically, then the authors should be more quantitative when discussing the model skill using metrics such as normalized mean bias, normalized mean error, etc. It is also interesting that the model over-estimates measurements, given that many air quality models fail to represent the high levels of $PM_{2.5}$ concentrations observed during peak episodes in Beijing owing to missing treatment of heterogenous chemistry, as described in several recent papers such as Wang et al. (PNAS, 2016, doi:10.1073/pnas.1616540113) and Cheng et al. (Science Advances, 2016, doi:10.1126/sciadv.1601530).

- Section 4.1: There are several species and sectors that have emissions that contribute to $PM_{2.5}$ formation. Which emissions are considered in the presentation of the results here? In other works, how is S_n defined? Are anthropogenic and natural sources included? What type of anthropogenic sources? Is it the total emission across all species? This is an essential missing detail. The results have little scientific or policy relevance in current form, given that they are only

presented in terms of local vs nonlocal sources (a point for which use of an adjoint model would be overkill).

- 8.9-11: This statement hasn't really been demonstrated. To use the adjoint sensitivities to "reproduce" the air pollution episode, one would need multiply the time series of sensitivities by the time series of emissions and show that their product matches the observations. This has not been done, nor would it likely work owing to nonlinearities. Claims of efficiency are also implied but not quantified. A single adjoint model integration is often several times (2 - 10) slower than a normal forward model integration. Thus what is the overall computational savings of their approach here over forwarfd methods, given the size of N and M, quantitatively?

- Fig 6(e) and (f) are good to know, but they are somewhat of a waste of an adjoint model. If the only interest was in the separation between "surrounding" vs "local" emissions of all $PM_{2.5}$ precursor emissions, this could have been achieved with only 3 forward model integrations (adjoint not needed). So the authors haven't really brought out the strength of their results to provide insight into spatial attributions beyond these two regions. Pie chart showing the influence by province, species, and sector would be much more interesting, and would start to approach a level of detail unobtainable without use of an adjoint model.

- Fig 8: Defining these ratios based on the area of the regions is not the best idea. It would be better to define the ratios based on the magnitude of the emissions in the different regions, since emissions intensity per unit area is not uniform.

- 10.25-28: Table 1 and the argument based on area isn't a great method, as discussed above. And I'm sorry but Table 2 and surrounding discussion just does not make much sense, and requires further clear explanation of what is being presented. What is the importance of the ration SC / PC? This needs to be explained. What are the percent values percentages of? Do these sum to 100% in some manner? Lastly, comparison to results of Zhai et al. (2016) appears to be

entirely qualitative, and no clear summary of how the two compare quantitatively is provided.

3 Minor comments

- 2.2: The first sentence is a bit vague, and should be clarified. Adjoint models are efficient for some types of sensitivity calculations, but not all. They are also efficient in terms of wall-time, but not necessarily in terms of memory or i/o.
- 2.14-19: This brief overview of "current" applications of adjoint modeling in atmospheric chemistry isn't a great fit for this paper, as it doesn't cover the first works in this area, historically, nor is it limited to only the latest works. Also, in attempting to cover all applications of adjoint model, the authors touch upon several areas (O3, CO, etc.) that aren't directly related to the topic of $PM_{2.5}$. I suggest the authors instead consider a more detailed overview of previous works, but one that is more narrowly limited in terms of scope, possibly to just sensitivity studies of $PM_{2.5}$.
- 3.2: There is a second paper by the same group using adjoint modeling to investigate sources of $PM_{2.5}$ in Beijing during the APEC period.
- 3.9: Could the authors clarify what is meant by "guidance on the enaction of dynamic environmental control policy"? What type of policy are they referring to (municipal? national? international?), and what is dynamic about such policy?
- 4.13: Technically a first-order finite difference calculation would require N+1 forward model integrations.
- 4.18: The theoretical equivalence of these approaches predates the work of Liu by many decades; I suggest the authors find a more fundamental reference. Also,

it is typical to only cite PhD thesis (as opposed to peer reviewed literature) when absolutely necessary, which is not the case here.

- 4.26: Adjoint sensitivities would only provide "exact" contributions for linear systems. However, $PM_{2.5}$ is formed nonlinearly, which needs to be addressed, or the interpretation and use of the adjoint sensitivities needs to be reconsidered.
- 5.14: "Unequilibrated" is not the correct word here. Nonlinear?
- Section 3.2: In addition to the physical processes treated in this aerosol model, please also briefly review what chemistry is included, both in the aerosol and gas-phase, and how the thermodynamic partitioning of species across phases is modeled.
- Section 3.3: Previous studies have shown that there are influences of emissions on $PM_{2.5}$ in your receptor cite from beyond the model domain considered here. Thus please explain how the influence from boundary conditions is tracked in the adjoint modeling.
- 7.12: This is a more correct interpretation of adjoint sensitivity results which should be considered in the earlier descriptions.
- Fig 5: It appears the emissions continue to spread by 72 hours of back integration. How then did the authors decide to stop the adjoint integration at 72 hrs? In other words, why did they not integrate backwards further in time? The lifetime of aerosols can be much longer than 3 days, so integration of back to a week to 10 days may be necessary to capture all non-local influences.
- Section 4.1: Please clearly define what is meant by "local" in this context. Is it just the single grid cell that contains the Beijing receptor cite?

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- 9.24-26: The non-local contributions do get small after 72 hrs, but as shown in Fig 6(d), the cumulative sensitivities have yet to asymptote to a constant value, which would indicate that sensitivities from early than 72 hrs may still play some role, although small. Also, sensitivities may have transferred to the boundary conditions, as mentioned previously.

4 Corrections

I started making grammatical corrections to the abstract, but stopped after only a few lines, as the entire manuscript needs substantial editing.

- 1.17: in detecting → to detect
- 1.20: south to → south of
- 1.21: at the south to → to the south of

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