Dear Referee,

Thank you very much for your recommendation for publication and your valuable comments. Answers and revisions of the manuscript are listed as follows:

1. Do you construct the adjoint model manually or automatically? If automatically, which automatic differentiation tool do you use?

Answer: We construct the adjoint model both manually and automatically. We will add related information in the manuscript in **Section 2.2.1** as follows:

In this study, the adjoint model was developed both manually and automatically. The Automatic Differentiation Engine, TAPENADE (Tangent and Adjoint PENultimate Automatic Differentiation Engine) (http://www-tapenade.inria.fr:8080/tapenade/index.jsp), developed at INRIA Sophia-Antipolis by the TROPICS team, was used to generate the tangent linear and adjoint code of sub-programs in the aerosol module CAM and the adjoint of corresponding interface programs. During the adjoint generation procedure, we should distinguish input variables from output variables and parameters. After that, manually assembly of the divided sub-programs and the interface programs, as well as validation of the tangent linear and the adjoint models were necessary.

In addition, the headline will be changed to "General introduction of adjoint theory and adjoint construction".

2. This paper aims at tracking influential BC emission regions. The following points may be added to its content: a) Typical adverse impacts of excess BC aerosol in the atmosphere: climate change, melting of glaciers, damage to plants, et al. (in Section 3); b) Explicitly explain what enlightenment can be drawn from BC sensitivity analysis. For example: can this adjoint methodology be applied on haze/$PM_{2.5}$ pollution control? Infer possible results of applying this adjoint sensitivity analysis method to SOA control.

Answer:

a) We will add typical adverse impacts of excess BC in the manuscript in **"Section 3"** as:

BC is the main light absorbing aerosol species; it alters the radiative properties of other aerosols with which it is mixed. In addition, it may also affect cloud formation and precipitation (Hakami, 2005), reduce crop production, decrease visibility, as well as harm human health. In one word, BC plays an essential role in atmospheric radiative forcing, climate change and air quality evaluation.

b) The following will be added in the "**Conclusion Section**":

Through analysing the result of BC adjoint sensitivity results, it's naturally to shine light on efficiently controlling haze using the adjoint method, after the development of the adjoint of all three modules in CUACE.

Possible results of SOAs emission sources adjoint sensitivity analysis inference:

SOAs are formed from both gases and particles, the non-linearity of SOA formation makes it more difficult to analyze compared with BC, who is 'emitted directly into the atmosphere predominantly during combustion'. We guess that the adjoint influential source regions of SOAs might be more deviated from the objective region, and more widespread. Moreover, the trends of adjoint sensitivity coefficients along inverse time series will surely be different from that of BC.

3. The description of the ajdoint model construction is not very clear. Section 2.1 presents that GRAPES-CUACE aerosol module involves six types of particles. If the adjoint of all these processes are constructed, then why only BC is analyzed in this research? Does this adjoint model include SOA adjoint processes? If not, besides explicitly explain it, a "future work" part might be added at the "conclusion" section as: Conclusions and Future Work.

Answer: Thank you very much for pointing this out. All processes in CAM (Canadian Aerosol Module) have been developed for their adjoints. However, the adjoint of the gaseous and thermodynamic equilibrium processes in CUACE are unavailable yet. We will add related information in the manuscript as:

Introduction Section:

In this research, the adjoint model of the GRAPES-CUACE aerosol module has been developed. As the adjoint of the gas-phase module and the thermodynamic equilibrium module are not available yet, only BC sources and primary sources of SOAs can be tracked. The newly constructed GRAPES-CUACE aerosol adjoint model was used in BC receptor-source sensitivity analysis to lay the foundation of further SOAs sources (primary and secondary) tracking as well as emissions inverse modelling.

Section 2.1:

The air quality forecasting system CUACE mainly comprises 3 modules: the aerosol module, the gas-phase module and the thermodynamic equilibrium module. CUACE adopted CAM (Canadian Aerosol Module) (Gong et al., 2003) as its aerosol module. In this research, we developed the adjoint of CAM.

Conclusion:

Through analyzing BC adjoint sensitivity results, it's naturally to shine light on designing efficient haze control schemes using the adjoint method.

4. In section 3.4, explain the reason for the sharp decrease in time series sensitivity coefficients (Fig. 8a) at 17-18 h ahead of the most polluted time point.

Answer: Wind field pattern plays an important role in air pollutants transport. Because the objective function is average BC concentration over Beijing at the highest concentration time point, and this high BC concentration is caused by emissions at upwind area, sensitivity coefficients at different time intervals correspond to different emission sources regions. As emission sources are unevenly distributed, sensitivity coefficients are influenced by wind direction and speed.

Through the analysis of 10-m horizontal wind field, this sharp decrease of sensitivity coefficients at 17-18 h ahead of the most polluted time point is due to change of wind field pattern at about 19:00 BT 3 July, 18 h ahead of the objective time point. At around 19:00 BT 3 July, relatively higher speed northwesterly wind to the north of BJ began to change to lower speed wind; meanwhile, to the south of BJ, lower speed southeasterly wind was changed to higher speed southerly wind. Therefore, before 19:00 BT 3 July, BJ was mainly affected by relatively clean northern Hebei region; after 19:00 BT 3 July, southern Hebei with intensive emission sources began to affect BJ. The different roles of south Hebei and north Hebei in affecting BJ air quality can also be reflected in the cumulative sensitivity coefficient distribution (Fig. 6).

5. The content in section 2.2.4 is too simple. Details have better be added, for example: what modification has been made to the standard model to facilitate the specific adjoint sensitivity analysis goals of the present study? Besides, move detailed captions in Figure 1 to this section.

Answer: We will revise **Section 2.2.4** in the manuscript as:

After each part and the assembled TLM and adjoint model have been verified, the GRAPES-CUACE aerosol adjoint model is constructed. The structures and parameters passing flowchart are shown in Fig. 1. In Fig. 1, ADJ is short for adjoint; Xn, Xn+1 represent model parameters after n, n+1 GRAPES-CUACE integral time steps, respectively; X^*_n , X^*_2 represent, correspondingly, X_n's adjoint $\partial J/\partial X_n$ and X_2's adjoint $\partial J/\partial X_2$, where J is the objective function; $\partial J/\partial X$ are forcing terms; structures and variables in solid line frames are related to forward simulation; structures and variables in dashed frames are adjoint simulation relevant. In addition, as GRAPES-CUACE is an on-line meteorological chemistry modelling system, the aerosol transport processes are extracted from GRAPES, therefore, a process called "aerosol-related transport adjoint" is in Fig.1.

When operating, the forward GRAPES-CUACE should be run first to save basic state values of un-equilibrated variables in checkpoint files. Intermediate values are recalculated or saved in stack during the adjoint integration. Then, the saved basic state values during the forward integration and the forcing terms are used as inputs in the adjoint backward simulation.

Other major revisions (minor changes are yellow highlighted in the manuscript):

1、 Subtitle of Section 2.2.1 is changed to "General introduction of adjoint theory and adjoint construction".

2、 Subtitle of Section 2.2.4 is changed to “Assembly and operation flow of GRAPES-CUACE aerosol adjoint”.

3、 Two references are added:

Hakami, A., Henze, D. K., and Seinfeld, J. H.: Adjoint inverse modelling of black carbon during the Asian Pacific Regional Aerosol Characterization Experiment, Journal of Geophysical Research, Vol. 110, D14301, doi: 10.1029/2004JD005671, 2005.

Seinfeld, J. H. and Pandis S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Second Edition, John Wiley & Sons, Inc., pp. 628-633.

Thank you for your comments and suggestions.