Reply to Anonymous Referee #3

This manuscript presents a modeling study to quantify the contribution of stabilized Criegee Intermediates (sCI) on the sulfate formation in the BTH region using a source-oriented WRF-Chem model. The topic is well within the ACP domain and is well addressed, the methodology is sound, and the results are well presented. The work can be accepted for publishing after the following minor issues are addressed.

1 Comment: The authors use the results of the sCI contribution on the sulfate formation at one station (NCNST) to draw a general conclusion in the BTH. This is a bit of overstretch. It needs to be clarified. Besides the results at the NCNST site, it would be valuable to select a larger source area and a downwind area to discuss the contributions of the sCI and other three pathways. With this expansion, the paper would present a more general picture of the sulfate sources in the BTH.
Response: We have clarified in Section 3.2: "*Figures 8b-e present the simulated temporal variations of the four pathways to the sulfate concentration averaged over the whole BTH from 04 to 15 July 2015. On average, the HR_SO_2 is still the dominant sulfate source, with a contribution of 35.3% (Figure 8b). The OH_SO_2 plays an important role in the sulfate formation, accounting for 33.1% of sulfate mass (Figure 8c). The primary emission pathway and sCI_SO_2 contributes 22.5% and 9.1% of the sulfate concentration, respectively (Figures 8d and 8e).*"

2 Comment: Regard the effects of sCI on the sulfate formation, in addition to the oxidation of SO2 by sCI, sCI may also contribute to the sulfate formation through enhancing the atmospheric oxidation capacity, since sCI may enhance the ozone formation and ultimately enhance OH, and thus enhance the SO2 oxidation by OH. The latter can be called the indirect effects by sCI. It would be helpful to evaluate the sCI's indirect effects.

Response: We have clarified in Section 4: "*Additionally, as a potentially important atmospheric oxidant, sCI may enhance ozone formation and ultimately the OH formation. The indirect effects of sCI on the sulfate formation by promoting the OH_SO_2 pathway should also be investigated further.*"

3 Comment: Not considering the aqueous SO2 oxidation in cloud or fog droplets on one hand may underestimate the sulfate formation, and on the other hand may overestimate the contributions from other sources, including the sCI source.
Response: We have clarified in Section 2.2: "*It is worth noting that the WRF-Chem model cannot well resolve clouds formed in the planetary boundary layer (PBL), so the aqueous SO_2 oxidation in cloud or fog droplets is not considered in the study, which might cause the sulfate underestimation, and on the other hand might overestimate the contributions of the four pathways to the sulfate concentration.*"

4 Comment: There is no information on emission inventory used in the simulation.
Response: We have clarified in Section 2.1: "*The anthropogenic emission inventory with a horizontal resolution of 6km is developed by Zhang et al. (2009), with the base year of 2013, including industry, transportation, power plant, residential and agriculture sources. The Model of Emissions of Gases and Aerosols from Nature (MEGAN) is used to calculate the biogenic emissions online (Guenther et al., 2006).*"

5 Comment: In the abstract, the statement "The primary emission accounts for around 22~24% of sulfate concentrations due to high SO2 emissions" is confusing. Is this due to the sulfate emissions and due to the SO2 emissions?
Response: We have revised the sentence in the abstract: "*The primary sulfate emission accounts for around 22~24% of the total sulfate concentration.*"

References:

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), Atmos. Chem. Phys., 6, 3181–3210, https://doi.org/10.5194/acp-6-3181-2006, 2006.

Zhang, Q., Streets, D. G., Carmichael, G. R., He, K., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J., Chen, D., Duan, L., Lei, Y.,Wang, L., and Yao, Z.: Asian emissions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys., 9, 5131–5153, https://doi.org/10.5194/acp-9-5131-2009, 2009.

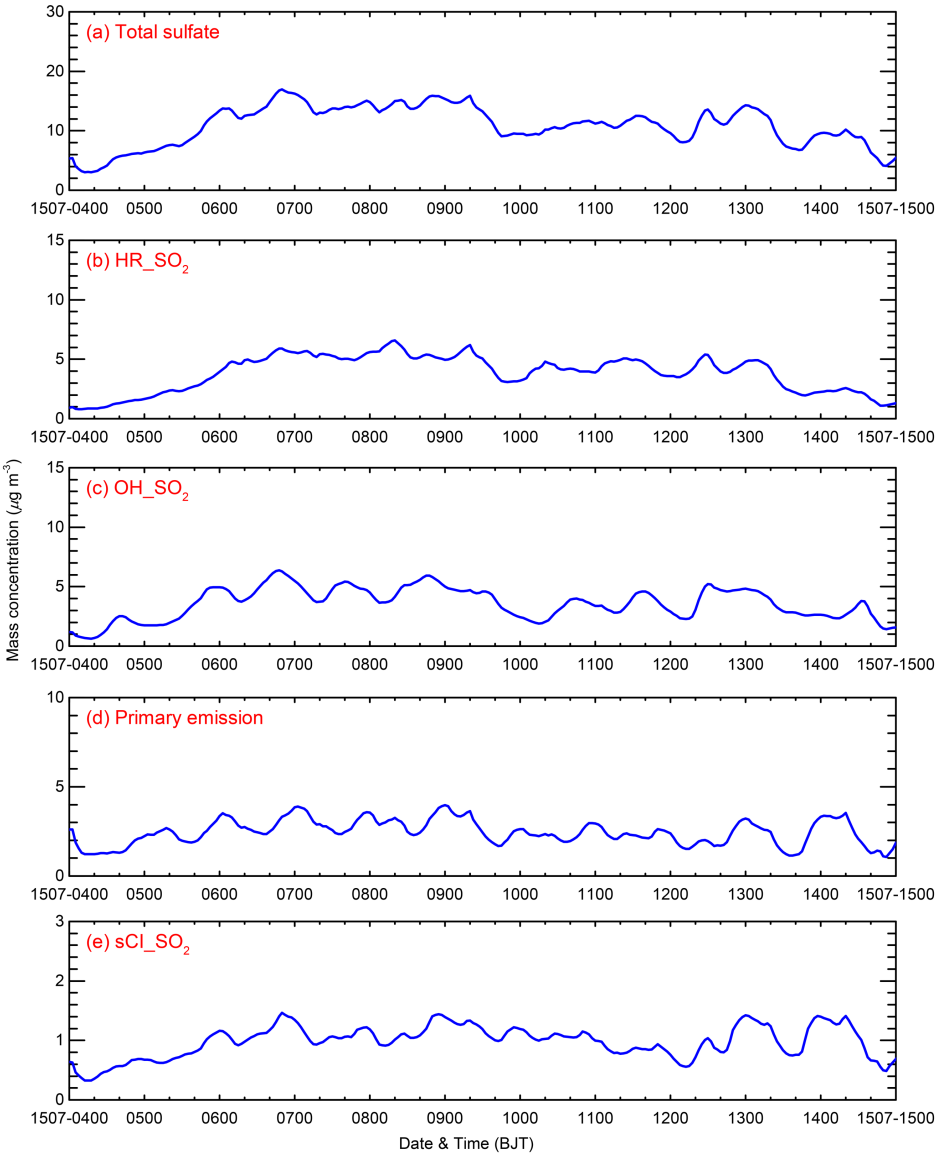

Figure 8 (a) Simulated diurnal profiles of total sulfate mass concentration, and contributions of the (b) HR_SO_2, (c) OH_SO_2, (d) primary emission, and (e) sCI_SO_2 to the sulfate concentration averaged over BTH from 04 to 15 July 2015.