Reply to Anonymous Referee #1

We thank the reviewer for the careful reading of the manuscript and helpful comments. We have revised the manuscript following the suggestion, as described below.

This paper presents WRF-Chem simulation results for a winter haze event in 9-26 January 2014 in Beijing-Tianjin-Hebei area. The results highlight the important effects of HONO, glyoxal, and methylglyoxal on SOA formation. The simulation results after considering all these effects yield significant improvement in comparison with observed SOA, O_3, HONO, HOA, BBOA, and CCOA. The results are significant and the presentation is in high quality. This reviewer only has a few minor concerns before recommending it for publication.

1 Comment: While the introduction provides a good literature review of SOA importance and recent advances in modeling SOA, it is a bit open ended because it didn't address why this paper is needed and how this paper differs from past studies.

Response: Previous studies have shown that CTMs are subject to underestimating SOA concentrations against measurements using the traditional two-product SOA module in China, particularly during wintertime haze days when the O_3 level is rather low. Therefore, in the study, we attempt to improve the SOA simulation based on a VBS SOA module, with focuses on the contribution of heterogeneous HONO sources and the uptake of glyoxal and methylglyoxal to the SOA formation during wintertime haze days in BTH. We have clarified in Section 1: "*Recent studies have demonstrated that CTMs are subject to underestimating SOA concentrations against measurements using the traditional two-product SOA module, particularly during wintertime haze days with rather low O_3 level (e.g., Jiang et al., 2012; Fu et al., 2012; Hu et al., 2017). Hence, it is imperative to improve the SOA simulations for supporting the design and implementation of emission control strategies to mitigate haze pollution in China.*

In the study, the VBS SOA approach with aging implemented in the WRF-CHEM model is used to attempt to improve the SOA simulation during wintertime haze days in BTH, with focuses on the contribution of the heterogeneous HONO sources and the uptake of glyoxal and methylglyoxal to the SOA formation."

2 Comment: It might be interesting to look at the difference in simulated vertical profiles of SOA and O_3. Early studies with field campaigns in ACE-Asia and TRACE-P showed the model's deficiency in simulating vertical profiles of SOAs. For this study, such analysis could be purely a model analysis because there was no observational counterpart to compare. But, still this can be an interesting and adds more 'meat' in the paper.

Response: We have clarified in Section 3.2: "*The vertical distribution is an important feature for evaluating the climatic impact of OA. Previous studies have shown large discrepancies between the simulated SOA vertical distribution and aircraft measurements (Heald et al., 2011; Tsigaridis et al., 2014). Although the OA vertical distribution measurement is not available during the simulation episode, analyses are still performed to explore the difference in simulated vertical profiles of POA and SOA, caused by the heterogeneous HONO sources. Figure 6a shows the vertical distribution of the average simulated POA and SOA concentration during the episode over IRSDE site in the BASE and HOMO case. POA and SOA concentrations decrease rapidly from the ground level to about 2 km, and are lower than 0.4 and 0.5 μg* m^{-3} *above 2 km, respectively. The POA concentration at the ground level is much higher than that of SOA, but its decrease in vertical direction is by far faster than that of SOA, which is consistent with the observation in Beijing by Sun et al. (2015). They have found that the SOA contribution to the OA mass at 260 m is higher than that at the ground level. The SOA enhancement due to the heterogeneous HONO sources is remarkable near the ground surface and rapidly decreases with the altitude, showing the dominant HONO contribution of the ground surface. Generally, the heterogeneous HONO sources do not substantially increase the* O_3 *concentration in the PBL, with an enhancement of less than 4% (Figure 6b).*"

3 Comment: Finally, is there any systematic diurnal variation of SOA? if so, how well model can simulate it? Diurnal variation of $PM_{2.5}$ has been of a high interest for air quality studies and such variation may differ by region. Recent studies show that in east Asia, WRF-Chem has deficiency to capture the observed diurnal variation (see citation below and references therein). It is worthy to add some analysis or discussion in the paper. To what extent the SOA simulation is improved as far as diurnal variation is concerned?

Lennartson, E. et al., 2018: Diurnal variation of aerosol optical depth and $PM_{2.5}$ in south Korea: a synthesis from AERONET, satellite (GOCI), KORUS-AQ observation, and WRF-Chem model, Atmospheric Chemistry and Physics, 18, 15125–15144.

Response: We have clarified in Section 3.2: "*Figure 5 presents the comparison of simulated SOA and observed OOA diurnal cycles averaged during the episode at IRSDE site. The observed SOA concentration continuously increases from the early morning (06:00 BJT) to the noon (12:00 BJT), due to the low PBL height and progressively increased photochemical production of SOA. After the noon, although the PBL commences to develop rapidly, the SOA concentration still increases until the evening (18:00 BJT), caused by the enhanced AOC to facilitate SOA formation. Compared to the HOMO case, the SOA diurnal cycle simulation is considerably improved in the BASE case against the measurement. The model with the heterogeneous HONO sources still fails to capture the observed SOA peak during the evening and overestimates SOA concentrations against the measurement from 00:00 to 06:00 BJT, showing the WRF-CHEM model deficiency in simulating diurnal variation of SOA formation (Lennartson et al., 2018). It is worth noting that the heavy haze pollution in Beijing is generally markedly influenced by the regional transport (Wu et al., 2017; Li et al., 2018), so uncertainties in the wind field simulations have large potentials to affect the SOA diurnal cycle simulation (Bei et al., 2017).*"

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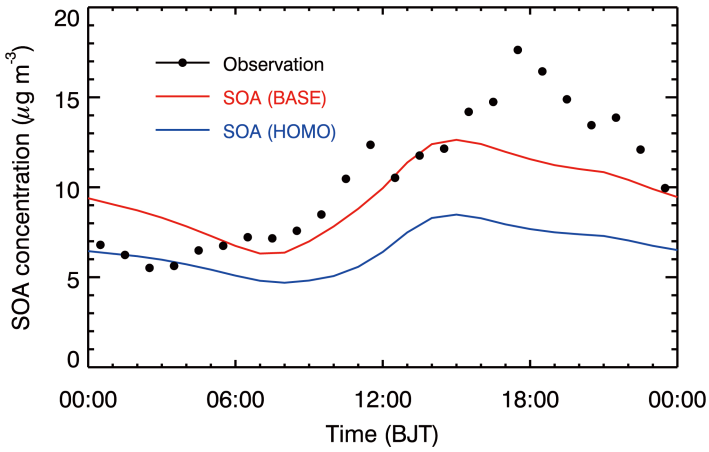

Figure 5 Observed (black dots) and modeled (red line: BASE case; blue line: HOMO case) SOA diurnal cycle averaged from 9 to 26 January 2014 at IRSDE site in Beijing.

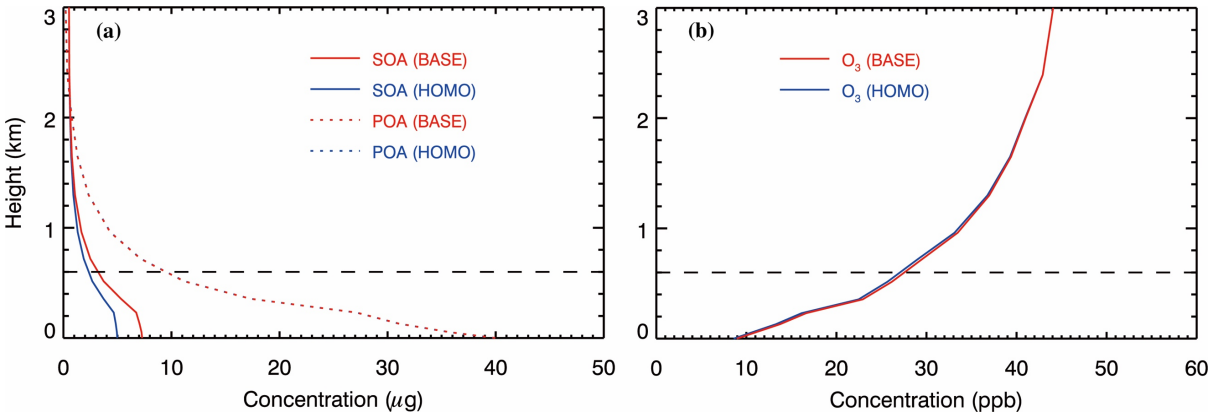

Figure 6 Vertical distribution of (a) SOA and POA and (b) O_3 concentrations averaged from 9 to 26 January 2014 at IRSDE site in Beijing. Red line: BASE case; Blue line: HOMO case.