Dear Dr. Nie,

I sent the revised version of your manuscript (Liu et al., 2019) to two experts (Referee #1 and Referee #3) for further review. Now I have received a report from Referee #3. Unfortunately, Referee #1 declined my invitation. In the first round of review, the major concerns from Referee #3 are about the calculation of OH concentration and the use of chemical reactions for OH production. I agree with the referee. Moreover, I think using simple parametrization of OH by $J(O^1D)$ does not fit and should be discouraged in this and other similar studies.

Equation (2) in the original version of the manuscript (referred to as E2_ori hereafter) was derived by Rohrer and Berresheim (2006) based on measurements at a GAW site in southern Germany. There is no doubt that such a strong correlation between OH and $J(O^1D)$ exists in the clean area (where $NO_x$ levels are low). It has been shown that the reaction $O(^1D)+H_2O$ dominates the global mean tropospheric source of OH (Lelieveld et al., 2016). However, E2_ori might not work well for the polluted area (where $NO_x$ levels are high).

Equation (2) in the revised version of the manuscript (referred to as E2_rev hereafter) was derived by Lu et al. (2012) based on measurements at a site in the Pearl River Delta. Lu et al. (2012) performed the fittings of measurement data without considering the variability of $NO_x$ and VOCs. E2_rev is an updated version of E2_ori with only a, b and c values changed, while the expression is functionally the same as that of E2_ori. It is doubtful if E2_rev can also be used to the Yangtze Delta. Actually, different a, b and c values were derived by fitting the OH and $J(O^1D)$ observed in urban Beijing (Lu et al., 2013).

Note that both E2_ori and E2_rev were used to investigate the correlation between observed OH concentration and $J(O^1D)$. There might be large biases in calculated OH from this parameterization especially under high NOx conditions, even for the same site where the measurements were carried out (e.g., E2_rev for the PRD site (Lu et al., 2012)). In contrast to the tropical and clean regions, the reaction $HO_2+NO$ (as well as $RO_2+NO$) contributes more to OH production than $O(^1D)+H_2O$ in the polluted areas, including eastern China (e.g., Lu et al., 2012;Ma et al., 2012;Tang et al., 2015;Lu et al., 2017). Variations of NO and VOCs, both of which influence the cycles among radicals (Lelieveld et al., 2008;Hofzumahaus et al., 2009), may have a large effect on the OH levels, which are not taken into account in this study.

This study shows that the photolysis of HONO (J(HONO)) has a larger contribution to primary production of OH than the effective photolysis of ozone ($J(O^1D)$), in consistent with the results from previous studies (e.g., An et al., 2009;Tang et al., 2015). However, I cannot see the role of HONO or J(HONO) in E2_rev. The OH calculated from E2_rev is used in several equations (e.g., Eq. 3). Should the result (e.g., $P_{OH}$(HONO) in Eq. 3) derived by using OH, which is primarily estimated from

E2_rev, have a strong feedback on the OH value? Moreover, could you explain why the second term (- $k_{NO+OH}$[NO][OH]) should be included on the right side of Eq. 3? I do not understand the meaning of $P_{unknown}$ in Eq. (10), some terms of which seems to be the same as the terms $L_{phot}$ or $L_{HONO+OH}$ in Eq. (9).

It would be very appreciated if these issues could be clarified before the acceptance of your manuscript.

Sincerely,

Jianzhong Ma

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