General Comments

Soni et al present new model results for air quality simulations that include the impacts of chlorine chemistry. The manuscript reports on updates to the chemical mechanism of CAABA/MECCA and discusses the impact of chlorine chemistry in two disparate regions (Leicester and Delhi). I have several general and specific comments that should be addressed prior to publication.

Response: Thank you for the constructive review; responding to these comments has improved our manuscript. Please find our responses below in blue fonts. The discussion added/updated in the manuscript is presented by red color font.

- The model reports a surprisingly large conversion of ClNO2 to Cl2. This is because of the large, condensed phase rate constant for ClNO2 + Cl- that was implemented in the model (Roberts et al 2008). More recent analyses have shown that this rate is likely too large. For example, the analysis of Haskins et al., JGR 2019, using field observations of ClNO2 and Cl2 suggested that this rate must be significantly smaller (of order 1E4 s-1). I think the authors should look at a sensitivity test to this rate to highlight that the selection of the ClNO2+Cl- rate constant has a really significant impact on the Cl production rate.

  Response: We thank the reviewer for raising this point and suggesting the article by Haskins et al., 2019. We agree that the aqueous phase rate constant of $ClNO_2$ + Cl^- reaction has significant effect on the production rate of Cl radicals as well as $ClNO_2$. As a consequence of including this reaction, the contribution of various reactions participating in Cl and $ClNO_2$ formation should change. In addition, significant changes could occur in concentrations of OH, HO_2, and RO_2 radicals in NEW simulation which contains the newly added Cl chemistry. These changes would also reflect in AOC over both the locations. In this regard, as suggested by reviewer a sensitivity test is performed with reaction rate of 5.7E4 mol^{-1} L s^{-1} and its effects are discussed by introducing a new section (4.4) in the revised manuscript. Figures S7 (simulated diurnal variations in Cl, $ClNO_2$, ClONO, OH, HO_2, and RO_2), S8 (Cl and $ClNO_2$ budget), and S9 (contribution of radicals in AOC) are added to the supplement to depict the changes occurred due to reaction rate of $ClNO_2$ + Cl^-. The above discussion is added in new section 4.4, and supplementary figures S7-9.

- As I understand the model treatment of heterogeneous and multiphase reactions here is quite different than what is in most models. Specifically, it appears that N2O5 is equilibrated between the gas and condensed phase using an equilibrium constant then permitted to react. It would be very helpful if the authors compared (and contrasted) this approach to the more common approach of using a reactive uptake coefficient for N2O5 chemistry that is sensitive to the chemical composition and phase of the aerosol particles. I expect that the two approaches would yield quite different results both with respect to magnitude and temporal trends. Since N2O5 chemistry is central to this study, this should be discussed.

  Response: In principle, employing a common approach for N_2O_5 chemistry using reactive uptake coefficients depending on chemical composition is expected to yield similar results, provided that the implementation of gammas and numerical integration is done properly. This approach essentially includes the rate coefficients from the fully

explicit kinetic model. In other words, the more common approach is a simple parameterization of the detailed aqueous-phase NO_2^+ chemistry that we have in our model and presented in the manuscript. It is worth noting that our model has the explicit reactions (forward and backward) for phase partitioning which often results in an effective phase partitioning equilibrium being established, consequently our approach is better compared to conventional approach. We do expect similar results with the two approaches but we think that such an analysis is beyond the scope of the present manuscript.

- Since the model has a comprehensive gas phase chemical mechanism, and the authors draw conclusions about the role of OH vs Cl radicals in VOC oxidation, it would be a nice opportunity to comment on the production of oVOC that stem directly (and only) from Cl+VOC reactions as they could be used in the future for testing the role of Cl chemistry. For example, what is the mixing ratio of chloroacetone?

Response: Cl reacts with hydrocarbons and acetone via H-abstraction. This does not form any Cl-containing molecules like chloroacetone. We don't have any such reactions in MECCA where the attacking Cl atom remains inside the organic molecule. In a future study, it could be checked if there are any important additions of Cl to a double bond which could form Cl-containing molecules. This discussion is now reflected in revised manuscript as outlook towards the end:

Lines 120-123: "In this study, Cl reacts with hydrocarbons and acetone via H-abstraction, and hence does not lead to the formation of any Cl-containing molecules, such as chloroacetone. This means that there are no such reactions in MECCA in which the Cl atom becomes part of the organic molecule. Therefore, for future research, it would be valuable to investigate the chemical kinetics of such reactions kinetics and their importance in the formation of organohalogen compounds."

Specific Comments

1. Line 55: This reaction is listed as H1, but H2 (and so on) are all Henry's Law Equilibriums. Should this be R1?

Response: H was currently used both for Henry's law equilibrium reactions and for heterogeneous non-equilibrium reactions. To avoid any confusions, we are now using "HET1" instead of "H1" in the revised manuscript (Lines 55-56).

2. Line 56: I would remove "recent studies" as it was shown 1997 by Behnke et al that the heterogeneous reaction of N2O5 could form ClNO2 on aqueous chloride containing films.

Response: We agree with the reviewer. The "recent studies" is removed and the sentence is updated   as:
Line 57: "However, N_2O_5 uptake on chloride-containing particles can produce $ClNO_2$ (Behnke et al., 1997; Thornton et al., 2010), ..."

3. Table 2: It would be helpful to add the Henry's Law constants (and references) to the table for the molecules studied. The Henry's Law constant for most of these gases have never been measured, thus the values reported in the literature are based on model fits to measured reactive uptake coefficients.

Response: We agree with the reviewer. The Henry's law constants with references are now added to Table 2.

4. Section 3: How is aerosol surface area and aerosol liquid water treated in the model? I appreciate that this may be described in one of the cited references (Rosanka?) But given its central importance to the science discussed here, I think it would be helpful if there was an explicit discussion.

Response: The following aerosol properties are defined in the model: radius, liquid water content, and chemical composition for both Delhi and Leicester, which are also mentioned in supplementary table S1. A line is added to the revised manuscript for clarification:

Line 151-153: "The values of aerosol properties (e.g., radius, liquid water content, and chemical composition) incorporated in the simulations for both Delhi and Leicester as provided in Table. S1."

5. Line 203: I think it would be helpful to cast the ClNO2 loss rates in units of per second as it is easier for readers to compare them to other locations.

Response: The $ClNO_2$ loss rates are defined in units of per second (molec cm^{-3} s^{-1}) in the manuscript. In addition, we have now also added the $ClNO_2$ loss rates in mol mol^{-1} s^{-1} in the text (Lines 253-257).

6. The ClNO2 + Cl- loss rate is enormous, and it would be helpful to see how that compares to other locations. Specifically, the ClNO2 uptake coefficient is quite small (< 1E-5) even on acidic aerosol. The surface area here must be enormous to drive a loss rate that is 10x that of photolysis (3hr lifetime). I think it would be very helpful to expand the discussion here to think more directly about this comparison.

Response: We are using alpha = 9.0E-3 from Fickert (doi:10.1021/JP983004N), not <1E-5.