We thank the reviewers for their time and comments. We have made efforts to improve the manuscript accordingly, please find response for corresponding points below.

Reviewer #1

This paper presents results describing the application of the GEOS-CHEM model to understanding the role of chlorine chemistry in the troposphere. The paper makes some new contributions and improves upon previous GEOS-CHEM modeling of halogen chemistry, but has so many major flaws and omissions that will require considerable modification. Since these changes may impact some of the findings, it is hard to know how to assess its publishability at this point. I have the following general and specific comments and questions.

General Comments:

There are number of errors and omissions in the reference list: papers that are noted in the text, but not in list, listed with errors, or just the wrong paper. Here is a list roughly in order of appearance: Liu et al., 2001 – which one, there are two in the reference list? Jaegle et al., 2012 – do you mean Jaegle 2011 here? Wesely 1989 is not in the list. Wang 1998 is not in the list. Jaegle et al., 2010 – do you mean Jaegle 2011 here? Martin et al., 2002 – do you mean 2003? Abbatt and Wachewsky, 1998 is not in the list Bey et al., 2001 is not in the list. Pszenny et al., 1991 – do you mean 1993 here? Roberts et al., 2008 is not in the list. Allen et al., 2007 is not in the list unless you mean Allan et al. 2011 Wang et al., 1994 is not in the list. Roberts et al. 2009 is not the correct reference, it should be this Roberts et al., 2009 (Roberts et al., 2009) Mielke et al., 2013 is not on the list.

We have corrected the reference list.

Chloride Sources: The paper ignores soil and wind-blown dust sources of particle chloride. Those were described by Sarwar et al., 2012 (Sarwar et al., 2012) (which by the way is not referenced in this paper) as the major sources of chloride in the Eastern U.S. and therefore will drive the chlorine budget in the middle of continents. This paper needs to give serious consideration to this source, and can use the IMPROVE network chloride data as a means to assess this continental source.

We now include model comparison to the IMPROVE data (Figure 5) and find that the marine source in the model can largely explain the observed chloride concentrations inland. We now cite Sarwar et al. (2012) for the fugitive dust source and point out that it would be small globally on page 5, line 14-17. We have added “global” to the title to emphasize that our focus is on the global scale.

Comparisons:

The selection of data sets for comparison was certainly not thorough, and obvious opportunities for comparison were apparently not considered. The paper seems to lean heavily on the Graedel and Keene work from 1995, but I believe further work has been done since then. There are comparisons that could be made that would greatly add to the model analysis and there are a number of data sets that were not included in the comparisons that were done.

Particle chloride can be compared to IMPROVE data (see for example Sarwar et al., 2012) to see how the model does. This is a long-term data set of considerable geographic extent.

To respond to the reviewer, we have added comparison to the IMPROVE data with a new Figure (Figure 5).

There are observations of HCl displacement by HNO3 in the LA Basin that need to be considered for comparison (Gard et al., 1998).

Observations in the LA Basin are not particularly relevant to us because of our coarse grid resolution (4^ox5^o).

There are a number of HCl data sets that were not compared with. Here is at least a partial list: Kim et al., INTEX-B (Kim et al., 2008), which includes one of the co-authors. CalNex ground site data for both particle chloride and gas phase HCl can be found at: https://www.esrl.noaa.gov/csd/projects/calnex/.

We do not include the INTEX-B measurements because Kim et al. (2008) was never published (it has remained in ACPD).

We included CalNex ship data (Crisp et al., 2014) data in Figure 4. These data are better suited for evaluating the model at the relevant resolution than the CalNex ground site.

There are a considerable number of ClNO2 data sets that were not compared with: Osthoff et al., 2009 – this covers essentially urban/industrial Houston and the near coastal environment. Sommariva et al., 2018 (Sommariva et al., 2018) covers several sites in England, including coastal sites. Le Breton et al., 2018 (Le Breton et al., 2018) Priestley et al., 2018. (Priestley et al., 2018) Tham et al., 2016 (Tham et al., 2013) Wang , Z. et al., (Wang et al., 2017b) Wang X., et al., 2017 (Wang et al., 2017a) Riedel et al, 2013 (Riedel et al., 2013) Mielke et al., 2015 (Mielke et al., 2015) Bannan et al., 2017 (Bannan et al., 2017) Young et al., 2012 (Young et al., 2012)(data set can be found at https://www.esrl.noaa.gov/csd/projects/calnex/) Zhou, et al., 2018 (Zhou, 2018)

To respond to the reviewer, we have added comparisons with Osthoff et al. (2008), Sommariva et al. (2018), Priestley et al. (2018), Mielke et al. (2015), Bannan et al. (2017), Jeong et al. (2018), Kim et al. (2014), and Tham et al. (2014) in Table 5. The comparison with Riedel et al. (2013)

was already included. Young et al. (2012) report the same dataset as Mielke et al. (2013) which is already included in the paper. Beyond that, Table 5 is already very long and as we now point out in the paper there is large representation error in trying to evaluate our global model with nighttime urban data (page 10, line 14-18). We also emphasize the global emphasis of our model evaluation at the beginning of Section 4.

It's true that there is only one lab study of the ClNO2 + Cl- Chemistry (Roberts et al., 2008), but the authors seem to have missed a key point of that paper: that reaction seems to only occur at pH=2 and below. Unless I missed something, this paper seems to allow this reaction after the alkalinity of the aerosol is neutralized. This may account for why the Cl2 is too high compared to the WINTER observations.

The reviewer did miss something. Table 2 gives $\gamma = 0$ for this reaction when $pH > 2$. But in any case, the aerosol pH reported by Guo et al. (2016) is smaller than 2 during the WINTER campaign and the same holds in GEOS-Chem. We have added more discussion in Section 4.2, page 12, line 9-15.

Specific Comments:

Page 10, Line 10, and Table 5. There are several data sets for which the model is a factor of 10 low, so it is hard to say the model performed "credibly". Also, most if not all of these data sets are available, so could be compared more in depth, not just maxima.

We now point out that there is large representation error in comparing our coarse-resolution model with nighttime urban data on page 10, line 14-18. More relevant is our detailed comparison to the $ClNO_2$ WINTER data in Section 4.2.

Page 11, lines 2&3. I believe the aircraft did missed approaches over airfields that could be used to do near-surface comparisons.

We deleted the sentence about lack of surface layer observations. These missed-approach data are not of much value to us considering the coarse resolution of the model.

Page 14, Line 22. Should be 'troposphere'.

Corrected (typo).

Reviewer #2

This paper by Wang et al. presents an extensive modelling study of chlorine chemistry using GEOS-Chem. The updates to the GEOS-Chem chemical mechanism are much needed. I think the paper is suitable for ACP, after the authors have addressed the following points.

MAJOR COMMENTS

As a general comment, I think it would be useful to have a more detailed discussion of the difference in the results between the version of GEOS-Chem with the previous mechanism and this new version. As it stands, the reader is mostly referred to previous papers. Figure 9 provides some of this information but it is presented as a sensitivity study, so it is not really a comparison of the old and new mechanisms.

We have added a supplementary figure S1 and a paragraph to Section 5.3 (page 15, line 25-34) comparing our simulation and GEOS-Chem standard version 11-02d.

Another general point is that the choice of datasets to compare with the model seems somewhat arbitrary. I understand it is not possible to use all the available datasets, but you have to explain why you selected certain datasets and not others for this work.

We now emphasize the global emphasis of our model evaluation at the beginning of Section 4 (and have added "global" in the title).We have added comparisons to the IMPROVE Cl^- data in new Figure 5 and to additional $ClNO_2$ datasets in Table 5.

On page 8: you say that the BrCl concentration in this work is lower than in previous modelling studies and you attribute this to a change in the chemical mechanism. Please add a note here that this point is further explained in section 5.2. Also note that BrCl measurements are discussed in the papers by Lawler et al. (see below). You may want to add that to the discussion in section 4.1.

We have added a note in Section 3 (page 8, line 26) that we would further discuss the lower BrCl in Section 5.2. We have also added the BrCl measurement in Lawler et al. (2009) to the discussion in Section 4.1, page 10, line 3-8.

On page 10 there is a brief discussion about the results by Lawler et al. (2010). First of all, it is either the 2009 or 2011 paper, please check which one you are referring to or add both. Both papers (2009 and 2011) reported high concentrations of HOCl and Cl2 when air was coming from continental Europe, not from Northern Africa as stated here. Please correct your statements. Those papers propose that high HOCl and Cl2 may be caused by aerosol acidification during long range transport and/or slower conversion of HOCl to Cl2 in the aqueous phase. These points are also investigated in Sommariva and von Glasow (2012),

which you may want to take into consideration. Is the high HOCl and Cl2 still unexplained in GEOS-Chem if you take into account the findings of these papers?

We thank the reviewer for raising this point. We have revised this part on page 10, line 3-13, including:

1) We were referring to Lawler et al. (2011). We now also add the data in Lawler et al. (2009) to the discussion.

2) We have corrected the statement for the sources of high HOCl/Cl_2 concentrations.

3) We have cited Sommariva and von Glasow (2012) and added it to the discussion. As Sommariva and von Glasow (2012) pointed out, a lower aerosol pH and/or slower rate for HOCl + Cl^- may explain the high HOCl but would decrease Cl_2.

On page 11: note that the results by Roberts et al. (2008) indicate that the reaction of ClNO2 + Cl- -> Cl2 is relevant only on very acidic aerosol. It may be that aerosol pH during the WINTER campaign was not low enough, which may explain why removing the reaction improves the agreement with the model. Can you please comment? Also, I think the reference is wrong in the bibliography.

We do consider this pH dependence and it is already listed in Table 2 ($\gamma = 0$ when pH > 2 for R7). In addition, the aerosol pH reported by Guo et al. (2016) are mostly smaller than 2 during WINTER campaign. We have added more discussion for this reaction in Section 4.2, page 12, line 9-15, and corrected the reference.

MINOR COMMENTS

Page 3: the title of 2.1 should be "GEOS"

Changed (typo).

Page 7, line 30: add space in "HCl +OH"

Added.

Page 12: is the lower Cl* in this work driven by lower BrCl, as mentioned on page 8?

The lower Cl* is driven by both the lower rate of Cl_y generation from acid displacement (as discussed in Section 3) and lower BrCl.

Table 5: there are more observations of ClNO2 available than those listed, especially in Europe and Asia. Either expand the table or explain why those were chosen. Also, it is not obvious in which order they are listed (year, season, region, concentration?).

We have added more datasets in our comparison of $ClNO_2$ in Table 5. We also add a statement at the beginning of Section 4 to emphasize that we only include data those not heavily affected by local anthropogenic sources. The order is based on latitude (from north to south), this is now explained in the Table footnotes.

Please tidy the references list, there are many mistakes.

The references list has been corrected.

Short Comments by McDuffie

1. The authors state on page 5, line 10 that anthropogenic sources of HCl were not included in their base case GEOS-Chem simulation. Although minor in a global sense, it is worth noting that Lee et al. (2018) reported observations of direct halogen (i.e. HCl, as well as Cl2, ClNO2, Br2, BrNO2, and BrCl) emissions from power plants sampled during the WINTER aircraft campaign. This is also important to note in Section 4.2 where model results are directly compared to WINTER chlorine observations.

We now include the power plant source of HCl following Lee et al. (2018) in a sensitivity simulation. This is discussed in Section 4.2, page 11, line 10-15.

2. Page 11, line 3, the authors state that WINTER aircraft observations did not extend to the surface layer. As also noted by one Reviewer, there were a series of missed approaches at airfields that could provide further vertical information.

We deleted the sentence about lack of surface layer observations. These missed-approach data are not of much value to us considering the coarse resolution of the model.

3. On page 10, Table 5 is referred to as a list of 'available' field observations of ClNO2. The GEOS-Chem simulations are then compared to these observations in Table 5 to evaluate the model performance. As noted by both Reviewers, however, there are many additional measurements of ClNO2 that were not included in Table 5. Measurements in addition to those provided by the Reviewers are from: (Edwards et al., 2013; Jeong et al., 2018; Kim et al., 2014; Liu et al., 2017; Osthoff et al., 2008; Phillips et al., 2016; Reyes-Villegas et al., 2018; Tham et al., 2018; Tham et al., 2014; Wang et al., 2018; Wang et al., 2016; Wang et al., 2014; Wild et al., 2016; Yun et al., 2018).

Thank you for pointing out these measurement data. We have added the comparisons with Osthoff et al. (2008), Sommariva et al. (2018), Priestley et al. (2018), Mielke et al. (2015), Bannan et al. (2017), Jeong et al. (2018), Kim et al. (2014), and Tham et al. (2014) in Table 5. Phillips et al. (2016) used the exactly same dataset as Phillips et al. (2012), which was already included in the paper. Wang et al. (2016) was also already included in the paper. Yun et al. (2018) used the exactly same dataset as Wang et al. (2016). There is no measurement of $ClNO_2$ concentration presented in Wang et al. (2014).

As pointed out in response to Reviewer 1 and in the revised text on page 10, line 14-18, comparison to nighttime urban $ClNO_2$ data is of limited interest for model evaluation because of the resolution mismatch. We also emphasize the global emphasis of our model evaluation at the beginning of Section 4.

4. The authors include the HOBr + Cl- ! BrCl reaction in their mechanism following Abbatt and Waschewsky (1998) and Fickert et al. (1999). They state in the discussion section on page 7 that the Cl source from this reaction is much higher than past simulations. Later in section 5.2, they also include discussion of previous work by Chen at al. (2017) who included a second HOBr + Cl- ! Br2 pathway that is dependent on the molar ratio of [Br-]/[Cl-], following Fickert et al. (1999). It is unclear in section 5.2 whether this additional pathway was included in the base case simulation here. If not, the authors should clearly state why it was excluded since the Fickert et al. (1999) laboratory work showed 90% Br2 formation from this reaction at ratios of [Br-]/[Cl-] typically found in ambient sea water. If included, this reaction would help reduce this Cl production pathway relative to previous simulations.

We have clarified on page 14, line 8 that we include this mechanism in our model simulation. We have also mentioned this mechanism follows Fickert et al. (1999) and would be further described in section 5.2 in the footnote of Table 2.

5. The authors have also included the direct reaction of ClNO2 with Cl- to form Cl2 (there is also mention of ClNO2+Br- on page 14, which should be added to Table 2. This reaction is thought to occur via heterogeneous uptake of gas-phase ClNO2 and further reaction with aqueous Cl- (or Br-). As noted by one Reviewer, however, it has been shown by Roberts et al. (2008) that this reaction only occurs at pH < 2 and should be limited here to only highly acidic aerosol. That said, the aerosol during WINTER were highly acidic (pH = -3 to 2) (Guo et al., 2016), which should activate this pathway. Even on these highly acidic aerosol, however, a recent study of WINTER ClNO2 yields by McDuffie et al. (2018a) reported that there was a negative correlation between particle acidity and CIMS observations of Cl2, which is opposite the trend expected from this reaction. In addition, there was no clear evidence in that study that gas-phase ClNO2 was being lost to heterogeneous processes (reaction with Cl or Br-). Since there is limited field data to support the presence of direct ClNO2 reactions in ambient aerosol, particularly during WINTER, the authors should consider eliminating direct ClNO2 reactions or provide further evidence to support their inclusion in this work.

Thank you for raising these points. We have added $ClNO_2 + Br^-$ as R8 in Table 2.

We do consider the pH dependence of $ClNO_2 + Cl^-$ and it is already listed in Table 2 ($\gamma = 0$ when pH > 2 for R7). We also add more discussion in this section on page 12, line 9-15 citing McDuffie et al. (2018).

6. The heterogeneous yield of ClNO2 is only mentioned in reaction R3 in Table 2, where it is defined using a laboratory-based parameterization from Bertram and Thornton (2009). This parameterization is used to predict both N2O5 uptake coefficient and ClNO2 yield. It is concerning that there is no discussion in this manuscript of the large uncertainties associated with these processes or parameterizations. First, this particular parameterization for N2O5 uptake does not consistently reproduce field-derived observations (e.g., Bertram et al., 2009;

McDuffie et al., 2018b; Wagner et al., 2013) and has been adjusted in recent GEOS-Chem simulations (Jaeglé et al., 2018; Shah et al., 2018) to better match nitrate observations during WINTER. While N2O5 is not the topic of this manuscript, this process directly impacts the net production of ClNO2, thus impacting the chlorine chemical mechanism and budget. The authors should therefore consider updating the N2O5 uptake parameterization in their simulations or discuss this as a source of uncertainty in their results. Second, this particular parameterization has over-predicted the ClNO2 production yield in every study that has compared its predictions to field-derived results (McDuffie et al., 2018a; Riedel et al., 2013; Ryder et al., 2015; Tham et al., 2018; Thornton et al., 2010; Wagner et al., 2013; Wang, Z. et al., 2017; Wang, X. et al., 2017). In addition, McDuffie et al. (2018a) recently found that the median WINTER ClNO2 production yield was over-predicted by at least 74% by the Bertram and Thornton (2009) parameterization. Since there are no field studies that support this parameterization as written in R3, the authors should adjust this reaction accordingly and discuss its uncertainties.

Jaeglé et al. (2018) and Shah et al. (2018) actually used the Bertram and Thornton (2009) scheme with no adjustment. Both papers updated the N_2O_5 uptake from old GEOS-Chem versions to the exactly same scheme as in our paper and found better match to the nitrate observations during WINTER. A major reason for the difference with McDuffie et al. is likely that GEOS-Chem treats reactive uptake by aerosols assuming an external mixture.

We have added more discussion for the N_2O_5 uptake parametrization and give our tentative explanation for the difference with McDuffie et al. on page 11 line 29 – page 12 line 4.