Response to Author Revisions

The authors have thoughtfully modified the manuscript to address many of the comments received during the review. There remain three sections that should be addressed prior to publication.

Page 11, Line 29

The authors have chosen to retain the $\phi(ClNO_2)$ parameterization based on previous laboratory results. The authors state that agreement of resulting simulated $ClNO_2$ with WINTER observations is in contrast with previous results (McDuffie et al., 2018a; Riedel et al., 2013; Wagner et al., 2013), that have shown the Bertram and Thornton (2009) parameterization over-predicts $ClNO_2$ production. Two minor points, 1) the cited references report that the *yield of $ClNO_2$* ($\phi(ClNO_2)$) is over-predicted by the parameterization, not necessarily the absolute amount of $ClNO_2$, 2) only McDuffie et al., 2018a ($ClNO_2$ yields) should be cited here as McDuffie et al., 2018b only reports $\gamma(N_2O_5)$ results from WINTER.

My bigger concern is that the authors have not sufficiently discussed the extent of the disagreement that exists in the literature between field and laboratory-derived $\phi(ClNO_2)$ results. I want to reiterate that while all laboratory-based studies have reported similar parametrizations of $\phi(ClNO_2)$ (Behnke et al., 1997; Bertram & Thornton, 2009; Roberts et al., 2009; Ryder et al., 2015), *every single* field study has shown that this $\phi(ClNO_2)$ parameterization over-predicts $\phi(ClNO_2)$ on ambient aerosol (see references in my initial review). There is currently no consensus on the source of this disagreement. In this analysis, it remains concerning that the authors have chosen to implement a parameterization that has been repeatedly shown to disagree with field results, especially without providing sufficient recognition of this disagreement or sufficient motivation for its inclusion.

Page 11, Line 33

The added discussion of $\gamma(N_2O_5)$ and $\phi(ClNO_2)$ results from (McDuffie et al., 2018a; McDuffie et al., 2018b) is useful here as these values were derived for the same campaign that is used to evaluate the model performance here. First, it would be better, in my opinion, to recognize the uncertainty in the $\gamma(N_2O_5)$ parameterization (as the authors have done), but then cite the $\gamma(N_2O_5)$ agreement between GEOS-Chem and the McDuffie et al. (2018b) box model that was previously presented in Jaeglé et al. (2018). As the authors have noted in their response, the $\gamma(N_2O_5)$ parameterization is the same here in Jaeglé et al. (2018) (though the particle chloride concentrations were calculated differently and Jaeglé et al. (2018)/Shah et al. (2018) also updated the organic aerosol uptake coefficient), so the reported agreement in $\gamma(N_2O_5)$ could help to validate the $\gamma(N_2O_5)$ calculation here.

My second comment is that the discussion of mixing state and chloride distributions across different aerosol types does not explain the overestimation of $\gamma(N_2O_5)$ or $\phi(ClNO_2)$ by Bertram and Thornton (2009) parameterization that was reported by (McDuffie et al., 2018a; McDuffie et al., 2018b), nor does it help validate their use here. For instance, since the $\gamma(N_2O_5)$ and $\phi(ClNO_2)$ parameterizations do not explicitly account of organic aerosol concentrations, $\gamma(N_2O_5)$ and $\phi(ClNO_2)$ would only be lower here relative to the McDuffie results if there were additional organic-associated chloride that was not being accounted for in GEOS-Chem by the assumption that chloride is only present in the SSA and SNA aerosol types. In addition, the authors may be further complicating this discussion by introducing the concept of mixing state because Bertram and Thornton (2009) explicitly state that their parameterization was 'designed to address an internally mixed particle population' (granted, SNA particles are assumed internally mixed in GEOS-Chem). Rather than discussing mixing state assumptions, I suggest simply citing and Jaeglé et al. (2018) to validate the $\gamma(N_2O_5)$ parameterization as described above.

Page 12, line 9 –
The discussion of the $ClNO_2 + Cl^- \rightarrow Cl_2$ reaction has greatly improved with the addition of the sensitivity test where R7 is removed. May I also suggest citing Fickert et al. (1998), Schweitzer et al. (1998), and Frenzel et al. (1998) who report $ClNO_2$ uptake coefficients for similar reactions with Br^- and I^- that are ~ 2 orders of magnitude lower than coefficients presented in Roberts et al. (2008). This could help to provide additional context for the magnitude of direct $ClNO_2$ uptake/reactions.

Minor Comments:
Page 7, line 7/8: Add reference to Roberts et al. (2008).
I would also suggest changing this sentence to: "The heterogeneous uptake of HOBr, HOCl, and $ClNO_2$, and further aqueous-phase reaction with Cl^- has been shown to be pH-dependent, with a higher efficiency in acidic solutions.

Page 7, line 8/9 -
Change to: "They are considered… , and the reaction of $ClNO_2$+ Cl^- further requires pH < 2, following laboratory results presented in Roberts et al. (2008)."

Page 9, line 25 –
remove the extra period at the end of the sentence.

Page 10, line 25 –
Change to: "…by an Iodide, Time of Flight, Chemical Ionization Mass Spectrometer (I^-TOF-CIMS).

Line 14 –
Suggest adding: "… is opposite of the trend expected from (R7), though there may be no trend on sufficiently acidic (i.e., pH < 2) aerosol.

References

Behnke, W., George, C., Scheer, V., & Zetzsch, C. (1997). Production and decay of $ClNO_2$ from the reaction of gaseous N_2O_5 with NaCl solution: Bulk and aerosol experiments. *Journal of Geophysical Research: Atmospheres, 102*(D3), 3795-3804. https://doi.org/10.1029/96JD03057

Bertram, T. H., & Thornton, J. A. (2009). Toward a general parameterization of N_2O_5 reactivity on aqueous particles: the competing effects of particle liquid water, nitrate and chloride. *Atmospheric Chemistry and Physics, 9*(21), 8351-8363. https://doi.org/10.5194/acp-9-8351-2009

Fickert, S., Helleis, F., Adams, J. W., Moortgat, G. K., & Crowley, J. N. (1998). Reactive uptake of $ClNO_2$ on aqueous bromide solutions. *Journal of Physical Chemistry A, 102*(52), 10689-10696. https://doi.org/10.1021/jp983004n

Frenzel, A., Scheer, V., Sikorski, R., George, C., Behnke, W., & Zetzsch, C. (1998). Heterogeneous Interconversion Reactions of $BrNO_2$, $ClNO_2$, Br_2, and Cl_2. *The Journal of Physical Chemistry A, 102*(8), 1329-1337. https://doi.org/10.1021/jp973044b

Jaeglé, L., Shah, V., Thornton, J. A., Lopez-Hilfiker, F. D., Lee, B. H., McDuffie, E. E., et al. (2018). Nitrogen Oxides Emissions, Chemistry, Deposition, and Export Over the Northeast United States During the WINTER Aircraft Campaign. *Journal of Geophysical Research: Atmospheres, 123*(21), 12,368-12,393. https://doi.org/10.1029/2018JD029133

McDuffie, E. E., Fibiger, D. L., Dubé, W. P., Lopez Hilfiker, F., Lee, B. H., Jaeglé, L., et al. (2018a). $ClNO_2$ Yields From Aircraft Measurements During the 2015 WINTER Campaign and Critical Evaluation of the Current Parameterization. *Journal of Geophysical Research: Atmospheres, 123*(22), 12,994-13,015. https://doi.org/10.1029/2018JD029358

McDuffie, E. E., Fibiger, D. L., Dubé, W. P., Lopez-Hilfiker, F., Lee, B. H., Thornton, J. A., et al. (2018b). Heterogeneous N_2O_5 uptake duirng winter: Aircraft measurements during the 2015 WINTER campaign and critical evaluation of current parameterizations. *Journal of Geophysical Research: Atmospheres, 123*(8), 4345-4372. https://doi.org/10.1002/2018JD028336

Riedel, T. P., Wagner, N. L., Dubé, W. P., Middlebrook, A. M., Young, C. J., Öztürk, F., et al. (2013). Chlorine activation within urban or power plant plumes: Vertically resolved $ClNO_2$ and Cl_2 measurements from a tall tower in a polluted continental setting. *Journal of Geophysical Research: Atmospheres, 118*(15), 8702-8715. https://doi.org/10.1002/jgrd.50637

Roberts, J. M., Osthoff, H. D., Brown, S. S., & Ravishankara, A. R. (2008). N_2O_5 oxidizes chloride to Cl_2 in acidic atmospheric aerosol. *Science, 321*(5892), 1059. https://doi.org/10.1126/science.1158777

Roberts, J. M., Osthoff, H. D., Brown, S. S., Ravishankara, A. R., Coffman, D., Quinn, P., & Bates, T. (2009). Laboratory studies of products of N_2O_5 uptake on Cl^- containing substrates. *Geophysical Research Letters, 36*(20), L20808. https://doi.org/10.1029/2009GL040448

Ryder, O. S., Campbell, N. R., Shaloski, M., Al-Mashat, H., Nathanson, G. M., & Bertram, T. H. (2015). Role of organics in regulating $ClNO_2$ production at the air–sea interface. *The Journal of Physical Chemistry A, 119*(31), 8519-8526. https://doi.org/10.1021/jp5129673

Schweitzer, F., Mirabel, P., & George, C. (1998). Multiphase Chemistry of N_2O_5, $ClNO_2$, and $BrNO_2$. *The Journal of Physical Chemistry A, 102*(22), 3942-3952. https://doi.org/10.1021/jp980748s

Shah, V., Jaeglé, L., Thornton, J. A., Lopez-Hilfiker, F. D., Lee, B. H., Schroder, J. C., et al. (2018). Chemical feedbacks weaken the wintertime response of particulate sulfate and nitrate to emissions reductions over the eastern United States. *Proceedings of the National Academy of Sciences*. https://doi.org/10.1073/pnas.1803295115

Wagner, N. L., Riedel, T. P., Young, C. J., Bahreini, R., Brock, C. A., Dubé, W. P., et al. (2013). N_2O_5 uptake coefficients and nocturnal NO_2 removal rates determined from ambient wintertime measurements. *Journal of Geophysical Research: Atmospheres, 118*(16), 9331-9350. https://doi.org/10.1002/jgrd.50653