Review of "Global tropospheric halogen (Cl, Br, I) chemistry and its impact on oxidants" by Wang *et al.*

General comment:

Wang *et al.* presented a description on tropospheric halogens (chlorine, bromine, and iodine) chemistry of an updated global chemical-transport model, GEOS-Chem, and assessed the effects of halogens on tropospheric oxidants and air pollutants. The paper has the potential to contribute to the increasingly recognized role of halogen chemistry in the troposphere. The topic of the manuscript also fits the scope of *Atmospheric Chemistry and Physics*. However, there are major concerns that should be addressed before it can be accepted for publication.

The biggest issue is the omission of anthropogenic (continental) source of reactive chlorine in the model, while there have been dozens of observations in the last decade suggested otherwise. Thornton et al. (2010) reported elevated levels of $ClNO_2$ at a continental site (~1400 km from the nearest coastline) in the U.S. Lee et al. (2018) observed high level of reactive chlorine species (HCl, Cl_2, $ClNO_2$, etc.) in the exhaust of coal-fired power plants in the U.S. Wang et al. (2016), Tham et al. (2016), Zhou et al. (2018), Yun et al. (2018), Peng et al. (2020), and many other recent studies in China consistently presented very high levels of $ClNO_2$ and other reactive chlorine species and almost all of these studies pointed to the anthropogenic source of chlorine. A recent report by Gunthe et al. (2021) suggests the existence of high loading of chloride in India.

The omission of anthropogenic chlorine resulted in many conclusions in the current manuscript that are not in line with previous observations, emission inventories, and model estimates which require further elaboration and/or adjustment.

(1) Line 66, a few emission inventories of anthropogenic have been proposed for China, including Liu et al. (2018), Fu et al. (2018), and Qiu et al. (2019). The anthropogenic chlorine in China alone could be up to ~0.5 Tg Cl a^{-1}, similar to the global biomass burning chlorine used here, so it's not "negligible". It's noteworthy that anthropogenic chlorine emission (mostly in the form of HCl, chloride) will be rapidly activated by anthropogenic NO_x and form reactive chlorine, e.g., $ClNO_2$, while in the vast open ocean, the HCl from the acid displacement mostly reacts with OH to relase Cl atom with a slow rate.

(2) Line 174-175, "Cl atom concentrations are usually highest along polluted coastlines", while including anthropogenic chlorine source might lead to a different answer. In fact, a few modeling studies, including one by the same authors as the current paper, have shown that anthropogenic chlorine leads to much higher levels of chlorine species over continental area than those along the coast, e.g., Hossaini et al. (2016), Li et al. (2016), Li et al. (2020), Wang et al. (2020), etc.

(3) Line 245, it appears that the authors did not consider the continental observations of HCl (e.g, the ones in Fig 7 in Hossaini et al., 2016) when conducting the model

performance evaluation. Based on Fig. 2 of the paper, I would expect that the simulated HCl over land would be much lower than the corresponding observations.

(4) Line 250, the same for $ClNO_2$. the authors only picked the observations at island and coastal environments, while the vast available measurements in China were not mentioned. An earlier version of GEOS-Chem with very similar chlorine source and chemistry setup (Table 5 in Wang et al., 2019), however, only simulated ~10% of the observed level in southern China.

(5) Line 257, this paragraph is particularly puzzling to me. Lee et al. (2018) specifically pointed out that the reactive chlorine species observed during the WINTER campaign are tied to the power plant plumes. However, the authors claimed that "modeled HCl is lower than the observations but mostly within the calibration uncertainty", while the current GEOS-Chem model did not include any power plant source of chlorine. Does it mean that the natural sources of chlorine used here is too efficient?

(6) Line 289, the co-existence of anthropogenic chlorine and VOCs emission means that the role of chlorine atom in VOCs oxidation would be more important than what is reported here.

(7) Line 303, "surface NOx increases over the continents and this is due to $ClNO_2$ chemistry". If $ClNO_2$ chemistry is an important factor, a full representation of its formation process (including the source of chloride) is then desired.

Specific comment:

1. Line 70-74, define 'long-lived' and 'short-lived'. I wonder why 'CH_2Cl_2' and '$CHCl_3$' are listed as long-lived species when they have a lifetime ~100 days. Is 'CH_2Cl' a typo? Also, '$CHBr_3$' has a lifetime of ~20 days, 'CH_2Br_2' ~130 days, and 'CH_3Br' ~ 2 years.

2. Reaction (1) to (4) and Line 120. What are the numbers used here?

3. Line 128, What was the original value?

4. Line 154, what is the simulation period? 2015 to 2016 with the first year discarded as spin-up? Please specify.

5. Line 167, how was '6.3' calculated? Also, the sum of 6.3 Tg (heterogeneous source) and 46 Tg (acid displacement) is different from 50 Tg (in line 76).

6. Line 179, what are the numbers in Sherwen et al. (2016b) and Zhu et al. (2019)?

7. Line 188 "HOBr is now more likely to react with S(IV)" is not consistent with line 190 "59% of HOBr heterogeneous reactions are with Br^- and Cl^-, and 41% are with S(IV)".

8. Line 195, It would be more informative if the distributions of SSA (both horizontal and vertical) are presented.

9. Please unify the ozone lifetime change in line 298 (‘10%’) and line 312 (‘11%’)

Technical comment:

10. Line 79, remove the extra ‘)’.

11. Line 152, please unify the use of ‘IONO’ or ‘INO_2’ and ‘IONO2’ or ‘INO_3’.

12. Line 165, ‘Cl*’ was defined in line 156.

13. Line 176, ‘global zonal mean’ should be ‘global mean’?

14. Line 271, ‘4.2’ should be ‘4.3’.

15. Fig 12, NO_x is not an oxidant. Also, the color scale should be improved for OH and NO_x.

Reference:

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