Review of the manuscript "Incorporation of multi-phase halogen chemistry into Community Multiscale Air Quality (CMAQ) model" by Kim et al., EGUsphere preprint repository, 2025.

The paper presents the implementation of halogen chemistry in the CMAQ model, including a comprehensive representation of chlorine, bromine and iodine sources and chemistry. Once the model updates are described, the study focus on reproducing ClNO2 and O3 observations over the Korean Peninsula during the KORUS-AQ campaign. The results focus on the improvements in the model-observation comparisons, including the index of agreement and other statistical parameters, particularly over two inland locations within a polluted / semi-polluted environment. Then, they identify the four main reactions in the model accounting for the largest fraction of the improvement. Based on this, the authors evaluate the influence of air quality in the whole modeled domain, highlighting the major and sometimes opposite differences observed over continental and oceanic domains, and provide general conclusions about the benefits of considering halogen chemistry in the study.

First of all, I would like to recognize the efforts from the group to implement their own version of halogen chemistry in CMAQ, which is of major importance as the community needs more modeling studies focused on the halogen influence on atmospheric chemistry and climate. However, I believe the current version of the work does not allow reaching a firm conclusion of the results obtained, probably because the paper attempts to address all at once the complete technical implementation, the observational improvements achieved over continental locations, and the overall impact and influences over different regions. While I leave for the authors to decide if it is convenient to present all of these developments within a single work or to partially split into independent companion papers, I recommend the authors to address the following major comments and submit a revised version for further consideration.

Major Comments:

P4,L93: In the introduction you clearly state that "We then investigate the formation of ozone using the new halogen processes", which is what the paper focus first … but then at the end the work attempts to provide much wider conclusions of the halogen influence on atmospheric chemistry and air quality. You may want to focus here on ozone production driven by ClNO2 chemistry, and leave the more general discussion for a companion paper.

P5,L111: Comparison with ClNO2 observations are of mayor importance for this study. Indeed, it might be worth to mention in the title. Current title and abstract give the impression of a general halogen chemistry development, while the work mostly focus on ClNO2 and its role for ozone production.

P20,L326-328: Your results for EXP_CAM are not surprising to me as the heterogeneous ClNO2 formation through N2O5 was not considered in Saiz-Lopez et al., (2014), which is your reference. However, it should be mentioned that those studies focused on oceanic and pristine conditions, and not polluted areas with high NOx and inland chlorine emissions. Indeed, further research from the group lead by Dr. Saiz-Lopez considered enhanced HCl and ClNO2 production within continental areas (see for example Li et al. 2022), which clearly showed important implications when anthropogenic chlorine sources are considered. Therefore I recommend including the complete chlorine scheme from CAM-Chem in your analysis to avoid reaching erroneous conclusion, or at least to clarify why your EXP_CAM simulations do not reproduce ClNO2 observations.

P22,L356-367: Of all four dominant process mentioned here, R20 in Table 1 (ClO + ClO) is particularly surprising to me, as this reaction is typically considered in stratospheric ozone depletion, but not for boundary layer studies. How do you explain that the surface observations are sensible to this reaction? Is it because of the subsequent Cl2 photolysis? Is due to the coupling of ClO + NO2? Can you explain how this reaction can contribute to ClNO2 formation during the night, when ClO abundance is zero and in addition, any Cl2 formed would not be photolyzed until the next morning?

P33,L547-562: I follow the explanation about the changes in the OH/HO2 partitioning, but oceanic SLH (particularly iodine) has been clearly shown to reduce the OH abundance (not increase it), and consequently to increase the CH4 burden and lifetime (Li et al., 2022). Your results over the ocean seems to contradict that. How could this be? Indeed, the null cycle mentioned in your work results in a shift of partitioning from OH to HO2 (which is fine), but the total OH abundance is controlled by O3 + hv --> O1D. Given that oceanic halogens reduce O3, there is less O1D and therefore OH formation should decrease. Have you discarded any influence from the BC affecting the overall results? Note that O3 changes for the EXP_Cl_Br_I - EXP_ctrl are in line with Li et al. (2022) for oceanic domains, but the OH changes are not consistent with the changes in O3 (unless I missed something).

General Comments:

P2,L41: put this values in the context of equivalent changes reported in the literature, here and elsewhere.

P3,L65: You could also cite other previous works with the implementation of halogen chemistry in WRF-Chem, e.g. from Badia et al., (2019). P4,L75: Similarly, given that you compare your results with those of the CAM-Chem model, you should also cite some of the CAM-Chem studies focused on the impact of halogen chemistry over the oceans as Saiz-Lopez et al., (2014, 2023), Iglesias-Suarez et al., (2020), Li et al., (2022).

P5,L106: What version of CMAQ did you consider? Have you used and/or compared w.r.t the previous implementation of halogen chemistry in CMAQ? (e.g. Sarwar 2015).

P7,L149: You should cite and compare your methodology and emission values for anthropogenic emissions with other works to put your regional results into context of the current literature. In P8,L180 you compare with Kim et al., 2023, but should also compare with the anthropogenic emissions from Saiz-Lopez et al., 2023.

P10,L206-211: How did you validate the overall halocarbon emission inventory implemented in your model? Based on the Ordoñez et al. (2012) inventory, global scaling of chl-a bitmaps was necessary to reproduce observations. In addition, what type of diurnal profile did you apply to the emissions?

P10,L212-218: Similarly, how did you exactly implement the SSA-dehalogenation process? Note this process is very efficient and depends on many parameters that present a large spatio-temporal variability (see Ordoñez et al. 2012 and Fernandez et al., 2014). Could you please provide more details about the implementation and the net bromine flux from sea-salt.

P10,L220-223: Halogen Chemical reactions. Please, provide at least a general introduction of which are the important reference works considered in this study.

P23,L380-385: In relation to the four highlighted reactions, I can think of many other processes that could be important to evaluate: for example: i) are Cl2 or any other species assumed to be uptake into the aerosol phase and provide Cl- (aq)? Ii) Did you consider any hourly variation in the emission strength of anthropogenic halogens of HCl and Cl2 (Eq. 1) that could impact your night-time results?. What about the NO2 sources that are required for the ClNO2

formation, how is their spatial and temporal variation? (this apply also to P33,L541). If all of these were found to be irrelevant, at least a couple of sentences explaining why they are not important should be given. I completely agree that further studies are necessary to investigate the main factors causing these discrepancies (which should be highlighted in the conclusions).

P26,L434-439: The way the text is written seems to indicate that this is a result from this study, while the opposite effect between continental (polluted) and oceanic (pristine) has been previously described in the literature (e.g., Li et al., 2022, Saiz-Lopez et al., 2023). Please rephrase to make it clear that your results are in agreement with those of previous studies.

P28,L462-464: this sentence makes no sense. Please rephrase. Are you sure the SSA dehalogenation process for bromine is well implemented in your model?. P33,L545: I would expect larger impacts of bromine than chlorine over the oceans, which is not the case. Could this be possible due to a small efficiency of the SSA-dehalogenation process for bromine?

P30,Eq.8 and Eq.9: Please control the F(Ox) expression for missing production channels and explain in case some terms are not considered. For the case of D(Ox) note that the Cl+O3 term should not be considered as it results in the formation of ClO, which is part of Ox (see Saiz-Lopez et al., 2014 for a complete list of all halogen-driven OddOx loss rates).

P36,L586-594: Once again, the conclusions concentrate on the importance of halogens to improve the model representation of ClNO2 observations, which are mostly related to air-quality in polluted environments. However, the final part of the paper focus on the wider implications of halogens over continental and oceanic domains, which were not validated before. Indeed, in L602-606 you summarize model results for several species but omit mentioning the inconsistent results found for OH. In case you decide to keep all the analysis in a single paper, a detailed discussion of this important issue should be included.

Language editing comments and Typos:

P2,L26 and elsewhere: Please refer to halogen chemistry, not chemistries.

P22,L365: It appears → it is evident.

P35,L583: replace I by I2 inside the parenthesis.

Table 2: Check for typos and consistency in R1 and R3.

Figures, Tables and Captions

Table 3: given the importance of reaction R6 in your results, more details should be given in the text. Note that most model implement different versions of R1.

Figure 4: it is not clear for me if this comparison exercise considers only nightime or 24-hs model output. Given that the model is shown to underestimate Cl2 observations during the day ... then it is expected that if 24-hs is considered the presented results would imply a night-time over-estimation for the EXP_CL_BR_I case. Am I right? Could you please clarify in the text?

References

Ordoñez et al., 2012 (https://acp.copernicus.org/articles/12/1423/2012/)

Fernandez et al., 2014 (https://acp.copernicus.org/articles/14/13391/2014/)

Saiz-Lopez et al., 2014 (https://acp.copernicus.org/articles/14/13119/2014/)

Badia et al., 2019 (https://acp.copernicus.org/articles/19/3161/2019/)

Iglesias-Suarez et al., 2020 (https://www.nature.com/articles/s41558-019-0675-6)

Li et al., 2022 (https://www.nature.com/articles/s41467-022-30456-8)

Saiz-Lopez et al., 2023 (https://www.nature.com/articles/s41586-023-06119-z)