Response to comments of Review 2

We thank the reviewer for the comments and suggestions on the manuscript. Our response (in blue) and the corresponding edits (in red) are shown below.

General comments

This paper presents a model study of the effects of halogen chemistry on the air quality of Europe. This study provides an interesting overview of the impact of halogens on ozone and other pollutants, a research question that is still open. The paper is well written and presented and I have only some minor comments (see below). Overall I think it is suitable for publication in ACP.

1. In Section 3.2 the CMAQ results are compared to the observations and to the GEOSChem results from Sherwen et al. (2017). First of all, there are other observations of ClNO2 in Europe besides those in Table 3. In fact some of these are mentioned in the Sherwen paper itself (as well as in Sommariva et al., 2018) and in Bannan et al. (2017). These measurements should be included in the discussion. Second, using the maximum observed concentration is not a good metric to assess the agreement with the model. For example, the observations in Phillips et al. (2012) show quite a range of peak nocturnal concentrations of ClNO2. I would also argue that GEOS-Chem shows better agreement with the measurements than CMAQ, especially wrt ClNO2 (lines 231-233). The discussion of the model-measurements comparison is better when dealing with iodine and bromine species, but please revise Section 3.2 to be more accurate.

Response: The observational results of $ClNO_2$ reported in Sherwen et al. (2017), Sommariva et al. (2018) and Bannan et al. (2017) have been included in Table 3 of the revised manuscript.

Table 3. The comparison of observed and simulated halogen species

Location	Species	Observation *	Simulation #
Hessen, Germany [a]	$ClNO_2$	800.0	273.4
London, United Kingdom [b]	$ClNO_2$	724.0	801.5
Weybourne, United Kingdom [c]	$ClNO_2$	65	373
Weybourne, United Kingdom [d]	$ClNO_2$	946	373
Weybourne, United Kingdom [e]	$ClNO_2$	1100 (summer) 75.6 (autumn) 733 (winter)	373
Leicester, United Kingdom [e]	$ClNO_2$	274 (spring) 74.2 (summer) 248 (winter)	274
Penlee Point , United Kingdom [e]	$ClNO_2$	922	319

Mace Head, Ireland [f]	BrO	6.5	10.1
Brittany, France [g]	BrO	7.5	0.4
Dead Sea [h]	BrO	100.0	0.2
Mace Head, Ireland [i]	IO	4.0~50.0	3.9
Brittany, France [j]	IO	7.7~30.0	1.1
Dagebull, Germany [k]	IO	2.0	9.0
Atlantic Ocean [l] (Prados-Roman et al., 2015)	IO	0.4 to 0.5 (daytime average)	0.4 to 2.0 (daytime average)

*: Maximum value (pptv).
#: Maximum value (pptv) from the HAL simulation.
a: Phillips et al. 2012.
b: Bannan et al., 2015.
c: Banna et al., 2017.
d: Sherwen et al., 2017.
e: Sommariva et al., 2018.
f: Saiz-Lopez et al., 2004.
g: Mahajan et al. 2009.
h: Matveev et al., 2001; Holla et al., 2015.
i: Allan et al., 2000; Commane et al., 2011.

Several field campaigns have been conducted in Weybourne in the past few years to measure $ClNO_2$. Sherwen et al. (2017) reported a peak concentration of 946 pptv. Bannan et al. (2019) reported a peak value of 65 pptv, and Sommariva et al. (2018) reported a peak value of 1100 pptv in summer, 75.6 pptv in autumn and 733 pptv in winter. CMAQ simulated a maximum of 373 pptv at that location while GEOS-Chem predicted 458 pptv. Sommariva et al. (2018) also reported measurements of $ClNO_2$ at Leicester with a maximum value of 274 pptv in spring, 74.2 pptv in summer, 248 pptv in winter, and that at Penlee Point a peak value of 922 pptv. CMAQ predicted a maximum of 274 pptv at Leicester and 319 pptv at Penlee Point. Eger et al. (2019) conducted shipborne observation of $ClNO_2$ in the Mediterranean Sea and reported up to 600 pptv $ClNO_2$ during their campaign, which is similar to the prediction of the present study.

We agree with the reviewer that the peak concentration of $ClNO_2$ (normally around the time of sunrise) at one location could have a large range, implying the large day-to-day variation of the level of $ClNO_2$ precursors (NO_x, O_3, Cl^-). The capability of CMAQ model to reproduce the maximum level of $ClNO_2$ at several locations throughout Europe (United Kingdom, Germany, Mediterranean Sea etc.) represents the ability of CMAQ to satisfactorily simulate emission, transport, and chemical transformation processes related to $ClNO_2$. Considering

that the present study was not designed to reproduce the level of $ClNO_2$ in a certain campaign, we think that the current validation metric adequate enough to show that the CMAQ model and the current setting can be used to investigate the halogen impact on the air quality in Europe.

We have removed the sentence that compares the performance of CMAQ with that of GEOS-Chem with regard to $ClNO_2$.

2. Figure 2 is interesting in the sense that it shows some different results from the corresponding figure 5 in the Sherwen paper especially when it comes to BrO. It looks like CMAQ is calculating lower concentrations than GEOS-Chem both for Cl and for HCl, which deserves some comment. It would also be good to include some of the European observations of HCl in this discussion. I realize that a comparison between CMAQ and GEOS-Chem is beyond the scope of this paper, but the differences in the geographical distributions of some species (and related impacts on O3 and other species) are sometimes striking and require at least a brief comment.

Response: CMAQ predicted lower level of BrO compared to GEOS-Chem. We have added a sentence to acknowledge that.

The predicted BrO levels over Europe are low (average value ~0.17 pptv) with the largest predicted value occurring within the Arctic circle while GEOS-Chem predicted >1.0 pptv level of BrO in Mediterranean Sea (Sherwen et al., 2017).

For the modeled level of Cl, CMAQ had a similar distribution and magnitude compared with the GEOS-Chem model, although CMAQ predicted a slightly higher maxima value (7.0×10^{-4} pptv, or $\sim 1.75 \times 10^{4}$ atom cm^{-3}) than the GEOS-Chem model (1.4×10^{4} atom cm^{-3}).

We have also added some discussion on the prediction of HCl level.

The observed level of HCl in Europe is in the range of <100 pptv to 5000 pptv (Hossaini et al., 2016 and the reference therein). The CMAQ model predicted a monthly average concentration of HCl between 6.3 and 1249 pptv, which is similar to the observation range. GEOS-Chem (Sherwen et al., 2017) predicted a maximum of 12 pptv for HCl, which is significantly lower than the available measurements in Europe.

In the original version, we compared the present study with the GEOS-Chem work in the simulation of halogen species ($ClNO_2$, BrO, and IO) oxidants (e.g., OH), and pollutants (e.g. O_3) and we also noted the difference between the two models. In the revised version, we have added some more discussions on the difference of the level and distribution of some species of the two models, e.g., BrO and HCl.

3. line 688: correct typo in name.

Response: Corrected. (Dub has been modified to be Dubé)