Comments on Long et al, ACPD
Anonymous Referee #3

Long et al present a unique global modeling study of the impact of halogen chemistry on tropospheric chemistry. Unlike previous model studies they explicitly model multiphase reactions in deliquesced aerosol particles. All previous global studies used a parameterized treatment of multiphase chemistry. Considerable effort and computing time have gone into these 10-year simulations. Overall, the results are very interesting but appear to (partly drastically) overestimate the impacts. This is likely caused by multiphase reaction cycles that are too efficient. This should have been pointed out and discussed more clearly and suggestions as to what the cause of this overestimate might be should have been made. It is surprising that the breakdown of organic bromine precursors was not included.
To me the real novelty of this study is to go beyond what has been done in previous studies in terms of process-level implementation of the underlying chemistry. However this should have been presented in a more critical way and discussed in more detail - and less optimistically - how this relates to observations. If presented somewhat differently I am sure that this study will serve to show in what direction research into the tropospheric impacts of halogens should go in the future.

Specific comments:
What is the actual reaction mechanism used in the model? This is never explained, the supplement doesn't contain a listing of the reactions that are used. This definitely needs to be included to understand and evaluate the model results.

Introduction: it is surprising that a brief overview of previous global model studies is missing. Several of these papers are not at all cited in the manuscript. To my knowledge the following are global model studies, that study the impacts of tropospheric halogen chemistry: von Glasow et al (2004), Lary et al (2005a,b), Yang et al (2005), Breider et al (2010), Saiz-Lopez et al (2012), Parella et al (2012).

p. 6071, l. 25: Lawler et al 2011 present unique data regarding chlorine speciation in the MBL. One of the current authors is co-author of this study, yet this paper was not mentioned here or below where the chlorine results are discussed.

p. 6073, l. 15-17: How are stratiform clouds treated?

Section 3: A discussion as to how modal-CAM-Chem (without halogens) would have been very helpful to assist the reader to put the results with halogen chemistry into context.

p. 6078, l. 2: You probably mean "upper troposphere" as this paper is not about the mesosphere and beyond...

p. 6079, l. 8: Delete “above the surface”.

p. 6079, l. 11: “moded” ◊ “modeled”

p. 6079, l. 15/16: I think this conclusion is a bit optimistic given that this is based on 2 studies only.

p. 6080, l. 12-18: While this explanation is plausible it would have been good to see some quantification or other support for it. Are there any field data that can be used to confirm this?

Tables 4/5: I’m very surprised about the emphasis on Br atoms. The reaction of Br + O3 is not a sink for odd oxygen which should be the metric used here. Most of the produced BrO photolyses back to Br and O3.

p. 6084, l. 15-25: Thornton et al discuss in depth the mismatch between measured particulate chloride and ClNO2 and argue that rapid repartitioning of gaseous HCl is the actual source for ClNO2. What is the chlorine source in the model? Does repartitioning play a role?

p. 6084, l. 28: I didn’t understand this adjustment.

p. 6085 Lawler et al (2011) presented unique data from Cape Verde regarding chlorine speciation. It is very surprising that this paper is not mentioned especially given that one of the current authors is co-author of this study.

p. 6087, l. 19-20: I don’t understand this statement/reaction sequence, please explain.

p. 6090: The discussion should include a discussion of ageing of sea salt aerosol and the related change in pH and hence in the importance of the reaction S(IV) + O3. This was explained in detail in Chameides and Stelson (1992, and other papers since, e.g. von Glasow 2006) and is key for the production of S(VI) in sea salt. Alexander et al (2005) included this very elegantly in a global study.

p. 6091, l. 1-5: I don’t quite understand this, please explain more. I don’t see how this difference should be responsible for the very pronounced differences between these studies.

General: The use of the word “deviations” is confusing. Please rephrase as “difference” or similar.

Table 3: Please explain the calculations in more detail/clarity, in the caption or in the text. Is “simulated” a 24h average? What does the ratio “day/night” show, is it the ratio of BrO at day to that at night?? How is the “estimated daytime mean” derived?

Figures 3 and 4 are too small.

References:

- There are a lot of mistakes in the references. Some papers are referenced in a different way in the text as in the bibliography. Saiz-Lopez et al 2012 is referred to interchangeably as Saiz-Lopez et al 2011 or 2012. Von Glasow et al (2004) is referred to as "von Glasow (2004)" and "von Glasow et al (2004)" in the text but in the bibliography a different paper is cited (von Glasow and Crutzen, 2004).