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Interactive comment on “Global stratospheric chlorine inventories for 2004–2009 from Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) measurements” by A. T. Brown et al.

Anonymous Referee #1

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This paper claims to present “Global stratospheric chlorine inventories for 2004–2009 from ACE-FTS measurements”. While the idea of a stratospheric chlorine inventory and its temporal evolution is to be welcomed I cannot recommend publication of this manuscript in its current form for the following reasons.

Firstly, I find the title is highly misleading as half of the inventory species come from a model simulation. Also, throughout most of the manuscript the discussion does not focus on “global stratospheric chlorine inventories” but on its trends including other

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regions such as the troposphere.

Secondly, and more importantly, I see this in large parts as an attempt to publish the same data set twice. ACE-FTS data of the same version (3.0) was already published two years ago in the Journal of Quantitative Spectroscopy & Radiative Transfer (Brown et al., 2011) for all measured chlorine species presented here. Brown et al., 2011 even covered a slightly longer period i.e. from 2004 to 2010. I can make out only two significant differences to the data presented in Brown et al., 2011:

1. Additional modelling data has been added for some minor species (e.g. CFC-115, ClO). These are partly replacing ACE-FTS measurements presented in Brown et al., 2011 which were found to be problematic (CFC-113, HCFC-141b, and HCFC-142b). Although the HCFC-22 retrieval used in this paper is new the paper discussing this new retrieval is still in preparation. So the new data here is either not from measurements or from an unpublished retrieval.

2. Unlike in Brown et al., 2011 the data presented here covers a wider latitude range and is also subdivided into latitude bands. The uncertainties in the data set are so large, that the validity of the presented annual decreases is already questionable. For this reason many recent studies have derived decreases over longer periods. This work goes even further by presenting total chlorine in different latitudinal bands. The authors however fail to demonstrate that ACE-FTS is capable to observe significant differences between these regions.

Thirdly, although this study tries to overcome measurement deficiencies by complementing it with modelled data it fails to include a major contributor to stratospheric chlorine and in particular its decrease: Methyl chloroform, which has accounted for a decrease of more than 45 pmol/mol of chlorine between 2004 and 2009, more than any other source gas. In addition, SLIMCAT has been extensively used to study short-lived source species. These contribute significantly to total chlorine and should be easy to include.

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I strongly encourage the authors to revise this work to improve conclusions and include more evidence for novel concepts, ideas, tools, or data. Below please find a number of specific comments.

Abstract Altitude ranges covered are not defined.

How can chlorine from halons dominate total chlorine at any altitude? Besides, H-1301 does not contain any chlorine.

If half of the species are modelled it would be worth pointing out to the reader whether the decreases derived from observations only were actually significant – in particular when arriving at the conclusion that the “Montreal Protocol has had a significant effect in reducing emissions of both ozone-depleting substances and greenhouse gases”.

“A positive slope with altitude” should be observed in the stratosphere as vertical transport is much slower there. But this is not known to a wide readership.

P 23492, I25 How are “stable chlorine-containing molecules” defined?

P 23493, I1-3 Has this been proven? If yes, please give a reference. Also, the “relatively small VMRs” of CH₃Cl are higher than those of the vast majority of species covered by this study.

P 23493, I4-5 Please give a reference for this statement.

P 23493, I5, 7 Both Molina and Rowland, 1974 and Farman et al., 1985 are not included in the reference list.

P 23493, I10 The Montreal Protocol does not prohibit use but controls production and consumption.

P 23493, I14-5 Please give a reference for this statement.

P 23493, I15-16 No predictions are included in this chapter of the WMO Ozone Assessment.

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P23494, I20-21 Please provide evidence for this statement. Is it because the largest number of molecules were included? Even if that is the case I would not consider a study that neglects methyl chloroform “most comprehensive”. And why is it superior to the multi-instrument study of Lary et al., 2007 which covers a much longer time period? Also, the sentence contains “budget” twice.

P23494, I23-25 This is a repetition from the introduction.

P 23495, I1 Brown et al., 2013 is nowhere to be found in the reference list. If this fluorine budget manuscript is not submitted yet, but has similar weaknesses, I advise the authors to consider the major concerns expressed above.

P 23495, I11 The instrument name should begin with capital letters.

P 23495, I25 According to Brown et al., 2011 and as pointed out later in this paragraph it measures 12 chlorine-containing species.

P23499, I19-20 How well do these volume mixing ratios compare to observations?

P23500, I3,5 Again, Brown et al., 2013 is nowhere to be found in the reference list.

P23500, I23-24 Altitude ranges over which was averaged are not defined.

P23500, I26 - P23501, I1 How was COCl₂ scaled? Also, how can you scale two species that come from different source gases?

P23501, I2-3 How does SLIMCAT compare to observations for HOCl and ClO?

P23501, I23-25 If total chlorine was derived between 0.5 and 53.5 kms the word “stratospheric” in the title is inappropriate.

P23503, I1-4 As recently shown by Hall et al. (AMTD, 2013) a flat 5 % error is not necessarily an overestimation of the error for the ground-based measurements.

P23503, I22-24 The concept of age-of-air in the stratosphere is rather complicated and not known by a broad readership.

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P23503, I24-26 Calling latitudes between 30 and 70 degrees “subtropical” is inappropriate. And if there is no “systematic difference between the slopes of the total chlorine” in the different latitude bands it only proves, that ACE-FTS and SLIMCAT are not able to resolve differences in vertical transport between the two bands described by Plumb, 2002.

P23504, I6-8 Only if there are no other biases influencing model or ACE-FTS results. It would be interesting to see, how organic and inorganic chlorine correlate over time given the title of the manuscript and also the exclusion of methyl chloroform and other species.

P23504, I16 This is not a “peak” but only the highest altitude considered in this study.

P23504, I19-20 Why is that interesting? And is this difference significant?

P23505, I9-11 Why is that rate of change larger? And is this in contradiction with the earlier statement, that “the difference between the morning and evening means is not statistically significant”.

P23507, I4 How was the tropopause defined?

P23508, I5-6 Which ODPs were used?

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