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Interactive comment on “ClOOCl photolysis at high solar zenith angles: analysis of the RECONCILE self-match flight” by O. Sumińska-Ebersoldt et al.

Anonymous Referee #1

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This study presents an analysis of ClO measurements from an aircraft flight performed in the Arctic stratosphere. The flight-route was planned in such a way that two measurements, separated in time, of the same air mass were obtained. A chemical Lagrangian model using various combinations of the kinetic parameters that control the effectiveness of the ClO dimer cycle was employed to reproduce the observed increase in ClO concentrations over sunrise. By using ClO measurements obtained before and directly after sunrise, together with model calculations, the authors provide an approach to constrain the photolysis frequency of ClOOCl, JClOOCl, independent of the ClOOCl formation rate constant k_{rec} . This is a good paper that complements existing field studies, as to date no field study has looked at the increase in ClO concentrations

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during sunrise and therefore was able to constrain JCIOOCI independent of krec. The paper presents new results that will be of interest to the readership of ACP. The paper would benefit from greater clarity in writing and from providing few more information on the match-technique used. After addressing my comments as stated below I recommend the paper to be published in ACP.

General

Neither the abstract nor the introduction provides any information about the measurements and measurement campaign e.g. where the ClO data were obtained from. I suggest adding one or two sentences (at least to the abstract) where you provide some more information on the measurements used, e.g. saying that it was an aircraft field campaign which was performed in Sweden in 2010 etc.

I think your abstract would benefit from providing some more information on what you actually did in this study and how you did it.

I noticed that you use abbreviations/acronyms without defining them throughout the paper. Please have a careful look through the whole paper and provide the definitions for the abbreviations/acronyms used e.g. chlorine monoxide (ClO) in abstract and introduction, polar stratospheric clouds (PSCs) in introduction, ozone (O₃), ClO_x, ppt etc.

Throughout the paper you are using the word ‘photolysis frequency’, ‘photolysis rate’, and ‘reaction rate’ and I think most of the time they are all referred to as JCIOOCI. Why do you use these different terms for JCIOOCI?

Minor comments

pg 18902 L3: ‘In the atmospherically relevant wavelength region. . .’ What wavelengths are included in the atmospherically important region? Please provide a range for the atmospherically relevant wavelengths you are referring to.

pg 18902 L3/4: ‘. . . published laboratory measurements of ClOOCl absorption cross

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sections and spectra are not in good agreement, resulting in significant discrepancies in JCIOOCI.' I think it would be better to say '... and spectra do not agree and so there remain significant discrepancies in JCIOOCI.'

pg 18902 L5: 'Previous investigations of the consistency with atmospheric observations of ClO and ClOOCl...' This sentence is not clear to me. The atmospheric observations are consistent with what (consistency of what with what)?? Please clarify.

pg 18902 L8: '... thus could only constrain the ratio of JCIOOCI over the rate constant of the ClO recombination reaction k_{rec} .' I think it would be better to say '... thus could only constrain the ratio of JCIOOCI and the ClOOCl formation rate constant k_{rec} .'

pg 18902 L10: I think it would be better to write 'ClO measurements' instead of 'ClO data'.

pg 18902 L 13: '...the rise in ClO concentration...' – Are you talking about the observed increase in ClO concentrations?? Please clarify.

pg 18902 L17: remove ',' after 420 nm, replace 'Additionally' with 'Furthermore', and remove 'the' before night-time ClO observations

pg 18902 L18: can not -> cannot

pg 18902 L23: 'ClO-dimer' or as on pg18903 L10 'ClO dimer'. This should be consistent throughout the manuscript. -> delete hyphen in 'ClO-dimer'.

pg 18902/18903: Say what M represents in reactions R1 and R3.

pg 18903 L9: I think it would be better to say '... i.e. the ClO dimer (ClOOCl) formation and thermal dissociation, ...'

pg 18904 L3-6: I think that there is far too much information in that sentence. I strongly recommend splitting this sentence in at least two.

pg 18905 L8: I don't quite get the second reason. Does that mean that all the other

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available absolute cross sections are associated with a different spectral shape? Perhaps some rewording could help to clarify this statement.

pg 18905 L9: Figure 1a shows...

pg 18905 L15 '... that governs the ClO dimer cycle' I think you should be more specific about what J actually governs. Is it the effectiveness of the cycle?

pg 18905: I assume the brackets '[]' denote concentrations, if so please say so. Later (Pg18909 last line) you write 'ClO rise' without using brackets. That should be consistent as I assume that in both cases you mean the increase in ClO concentrations. Is that correct? And again, are you talking about the observed increase in ClO concentrations?

pg 18905 L21: Does the reader at this point now what you mean by 'so called self-match flight' and 'self-match flight pattern'? Why is it a self-math flight? You should include some words about the match technique as this is the first time you use these terms.

pg 18905 L24: 'the sunrise' -> remove 'the'

pg 18906 Sect 2: I think that section should be included in Sect. 3.3 of this paper. It seems to be a bit random here and for me it is not clear why you are talking about the cross sections right here. If you leave that section here, please clarify why this is the best place and you might want to think about a different title for that Sect. 2?

pg 18906 L1: This is not a good starting sentence for Sect. 2. At this point, it is not clear to me why you mention these four sets of ClOOCl absorption cross sections/scaled spectra now and how you are going to use them in your study. I would like to see a few more details in an introductory sentence with referring to later sections or as mention above merge this Sect. with Sect. 3.3.

pg 18906 L21: 'Burkholder et al. results generally...' I think it would be better to write: 'The results presented by Burkholder et al. generally ...'

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pg 18907 L4: Please add 'HALOE (Halogen Occultation Experiment)'. Why now using 'O3' when you so far always used 'ozone'.

pg 18907 L5: The explanation for CLaMs comes in section 3.3 but really should be included here.

pg 18907 L7: Is the word 'rises' the right word to use or would be 'ClO concentrations' better? I'm asking because it is not clear to me what 'rises' you mean. I assume that you mean the ClO increase as soon as sunlight is available to from ClO via R2. Perhaps just rewording might help to clarify this statement. How about: 'To compare the ClO concentrations resulting from the considered photolysis rates (i-iv) with observed ClO concentrations obtained from the self-match flight. . .' Please clarify.

pg 18907 Sect 3.1: Again, you need to say some more words about the match technique and refer to one or two papers. I'm not sure that all readers will be familiar with the match technique and the related terms you are using here. First time you mention 'match-pair trajectories'. You should also add some more words what you mean by saying this.

pg 18907 L20-25: To clarify what you did, I strongly recommend that you split that long sentence into at least two sentences. Furthermore, you should be more specific what you mean by saying 'matching measurement'. In addressing my comment above this might not be necessary.

pg 18907 L25: Not clear what 'This' refers to.

Sect 3.1: I recommend to add a few sentences that describe a bit more clearly what you did in terms of obtaining the ClO measurements, i.e. describe in more detail the match technique and the trajectory calculations.

pg 18908 L26: Why is it important for your study to describe the COPAS measurements in such details? Are these details really necessary here? I assume that this description belongs to the paragraph 'To examine the accuracy of the matches. . .' and so I would

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combine the two paragraphs between L21 and L7 (pg18909).

pg 18909 L4: 'an other' -> 'another'

pg 18909 L6: 'In the COPAS data, the exhaust gases of Geophysica were observed several times on the inbound flight leg confirming the good performance of the self-match flight.' You repeat that statement later in section 4.1.

pg 18909 L13: 'wind fields and temperatures' -> 'wind and temperature fields.'?

pg 18909 L22: 'where' -> 'were'

pg 18909 L26/27: '15 h' -> '15 hours'

pg 18909 last sentence: I think you should be more explicit. Say that 196.6 K lies above the temperature threshold for heterogeneous reactions to occur and therefore the contribution to the ClO increase from Cl₂ release can be neglected.

pg 18910 L7: It is not clear to me what you are referring to when writing 'match radii'. How is a match radius defined? Please clarify.

pg 18910 L8: '...potential temperature...' -> '...potential temperature (θ)...'

pg 18910: You really should say what ' $\Delta\theta$ ' refers to (is defined as)? You might want to consider rewording that sentence.

pg 18910: 'To further test the accuracy of the matches, observations of the long-lived tracer N₂O by the HAGAR instrument were used.' You said that before. And you also use COPAS observations to test the accuracy of the matches. Can you avoid this repetition?

pg 18910 L23: '... and taken from ..' -> '...and Keq taken from...'

pg 18910 last line: '... from the night-time ClO measurements...' -> '... from the night-time HALOX ClO measurements...'

pg 18911 L7: 'Only the Keq from ...' -> 'Only Keq from...' or 'Only the Keq value from C7940

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...'
pg 18911 L9: 'Therefore in our study... for the further analysis.' -> 'Therefore, in our study, ... for further analysis.'

pg18911 L25: Not clear what 'This point...' refers to.

pg 18912 L11: What is the 'theoretical line'? The calculated/modelled ClO concentrations?

pg 18912 L13: '... calculated for i) (scaled absorption...)' -> remove ()

pg 18913 L2: Why did you use krec from JPL09 and Nickolaisen et al. 1994 and not from e.g. Troler et al. 1990, Bloss et al. 2001 etc.? What was the basis for the choice of those values?

pg 18913 L13: 'At these relatively low SZAs...' What SZAs are you referring to?

pg 18913 the sentence: '... parametrisation underestimating...' -> parameterisation

pg 18913 L27: remove 'too'

pg 18914 L14: Pope et al. was also scaled to Lien et al. in the same way as von Hobe et al. You should then mention it for von Hobe et al. and Pope et al. and not only for von Hobe et al.

What implication do your results have for modelling ozone loss in polar regions as current model studies mostly use the JPL recommendations for the kinetic reaction rates?

Figures

Fig 1: I think that the colour scale is inappropriate. Panel (a): choose a different colour for Cox and Hayman, as it is not readable at all in a printout. Also the absorption spectrum from Huder & De More is not recognisable in that panel. Please make sure all lines are clearly visible and all labels are large enough. Panel (c): It is very hard to

see the lines and to distinguish between the dotted red and dotted orange line. Which line represents $I(\lambda)$ and what is represented by the dashed and dotted lines?

Fig 2: 'of the flight', delete 'the'.

Fig 3: 'panels' -> 'panel', remove 's'; Say what is represented by the large circles. In the lower panel the colour scale is again not clear. It is very hard to see the blue line. Is it necessary to show all three coloured lines?

Fig 4: The label on y-axis should read 'ClOx mixing ratios, ppt'. You take Keq from Ferracci & Rowley, from JPL09, and from Plenge et al., so you should write 'Keq: ...' instead of 'ClOx: ...' (left corner of Fig 4). '...in combination with various Keq parameterisations.' -> You used three Keq values so why not just saying it: '...in combination with three chosen Keq...'?

Fig 5: Your y-axis reads 'Reaction rate...' which is the photolysis rate, right? So why not label the y-axis with 'photolysis rate, 10^{-3} s^{-1} ? - replace 'in dependence of' with 'versus'

Fig 6: Your y-axis should be labelled with: '[ClO], 109 molecules cm^{-3} ' - '...simulation employing photolysis rates J_{ClOOCl} based on i-iv' -> 'simulations using photolysis rates J_{ClOOCl} based on i-iv (solid lines).'

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 18901, 2011.

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