Manuscript prepared for Atmos. Chem. Phys. Discuss.
with version 2014/09/16 7.15 Copernicus papers of the LaTeX class copernicus.cls.
Date: 10 August 2015

Author response to reviews of: A Match-based approach to the estimation of polar stratospheric ozone loss using Aura Microwave Limb Sounder observations

N. J. Livesey[1], M. L. Santee[1], and G. L. Manney[2,3]

[1]Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California, USA
[2]NorthWest Research Associates, Socorro, NM, USA
[3]New Mexico Institute of Mining and Technology, Socorro, NM, USA

Correspondence to: N. J. Livesey, (Nathaniel.J.Livesey@jpl.nasa.gov)

Abstract

The Copernicus LaTeX style files demand an abstract, so here it is.

1 Introduction

We thank the reviewers for their helpful comments on the discussion paper. Detailed responses are given below. Reviewer comments are quoted in italics; our responses are in upright font.

2 Response to reviewer #1

This paper extends the Match technique from sonde observations to global MLS satellite data. In the Match technique, pairs of ozone observations that are connected by an air mass trajectory are combined to a global ozone loss estimate.
The paper is well written and is a valuable addition to the scientific literature. With the use of the MLS data set, the method largely benefits from the huge amount of data, although the single data profile is less accurate than an ozone sonde. With this method, the authors are able to provide much more detailed statements about chemical ozone loss than the "classical" ozone sonde Match.
Especially, the additional use of N_2O *for separately deriving the transport error is significantly strengthening the method. Also the sensitivity with respect to various assumptions (Fig. 2) is valuable. I would recommend it for publication in ACP, although I have a few suggestions summarized below.*

We thank the reviewer for their positive comments and succinct summary of the work described in this paper.

p. 10050 / fig 1. Figure 1 shows an example of the data that contribute to one Match result. With MLS data, there are orders of magnitude more matching pairs per data point than in the sonde Match analysis. This gives much better statistics. However, the evaluation is chosen such, that the fitted line includes the origin (0,0). In turn this means, that the weight of individual match is larger, if the sunlight hours are larger. In the shown example, the data with more than 100 hours likely have larger error in trajectory location. By eye, it looks like if the data were restricted e.g. to 60 sunlight hours, the slope would be steeper. A sensitivity with respect to a "cutoff maximum sunlight time" or "minimum matches per hour/20ppb bin" would be interesting. This is similar, but not identical to the shown sensitivity with respect to the match trajectory length.

Forcing the ozone loss fit to pass through the origin (0,0) is a standard part of the Match approach, and has been shown to give rise to a statistically unbiased estimate of ozone loss (e.g., the Lehmann reference in the manuscript).
We have implemented the "cutoff maximum sunlight time" sensitivity test described by the reviewer and found, as they and we expected, similar results to those seen when limiting the total "duration" of the Matches. Specifically, for limits of 70 hours or longer the estimated loss rate vs. number of Matches curve was very close to that of the "total lifetime" tests. The 70 hours case reduced the number of matches from $\sim$15,000 to $\sim$12,000. More stringent limits reduced the loss estimate by about twice the amount found in the total lifetime case (for a given number of matches, i.e., x-coordinate in Figure 2a). Note that, contrary to the reviewer's expectations, the slope got shallower, not steeper. To avoid further complicating Figure 2 (which reviewer #3 already considers somewhat cluttered), we have not added an additional line in the plots for this criterion; rather, we have opted to mention these findings in the text.
Specifically we have modified the text as follows (additions in italics):

> To check whether it is simply the mean match duration that is the underlying driver of this sensitivity (i.e., whether having less elapsed time between matched observation pairs inherently leads to larger estimates of ozone loss rate, regard-

> less of the amount of sPV variability), an additional set of perturbations was performed where the match ensemble was randomly downsampled in a manner favoring retention of matches with durations shorter than a given threshold (lavender colored points and line). This random thinning, while reducing the number of matches and the mean match duration as intended, has little impact on the estimated loss until the number of matches is reduced by around 40 % (similar behavior is seen in the flank divergence case*, as well as when we instead eliminate airmasses exposed to the sun for more than a given amount of time between observations, not shown*). The sPV criterion, by contrast. . .

This test is preferable to the alternative "minimum matches per hour/20 pbbv bin" approach also suggested by the reviewer because the binning required for that approach is not part of the fit algorithm; it was implemented solely for the purposes of drawing Figure 1.

---

p. 10056f, section 3.5. constraints on nighttime ozone loss. Nighttime ozone loss can be shown to be negligible in certain regions/periods and is not to be discriminated in other regions. Although it is currently not believed that there is nighttime ozone loss and it had been shown by the bi-variate analysis by Rex et al., that nighttime ozone loss is not likely, I have the feeling, that more could be concluded from this analysis. Do I understand this correctly, that one would get no nighttime ozone loss in some cases and not enough accuracy to discriminate in other circumstances? I suppose that there is no period/region with a statistically significant derived nighttime ozone loss. If this is the case, I think that it would be valuable to show these regions in which there is no significant nighttime ozone loss, as a "partial" proof.

We thank the reviewer for this thoughtful comment. Despite looking into this issue further in the light of this comment, we feel unable to strengthen our position on this issue in the paper. As discussed in the manuscript, the large density of MLS observations means that noise in the MLS measurements has negligible impact on estimated loss. The major influences on our results are thus systematic errors in the trajectory calculations and the choices of criteria/threshold etc. discussed in sections 3.2 and 3.4. This presents a challenge to properly assessing the "significance" of estimated day/night partitioning of ozone loss. We feel that developing a more robust approach to this quantification, likely including some of the techniques discussed in our reply to the reviewer's next point (see below), is beyond the scope of this paper. Accordingly, we prefer to leave this discussion in the manuscript unchanged.

Variability of the results. As said in the paper, the variability of ozone loss in the different studies is indicative of the individual methods (data locations and time, vortex edge definitions). It results from the wish to deriving one single average number for a quantity that is variable, especially as a function of equivalent latitude. The high statistics of the matches shown, could also be used to highlight this point, i.e. to show the ozone loss rate and/or the accumulated ozone loss sub-sampled as a function of equivalent latitude or sPV bins and time.

We agree that this is logical next step for this analysis. However, it is best performed using a "multivariate fit" approach, simultaneously estimating loss rates in different equivalent latitude, altitude and time regions. This approach has been developed and is undergoing testing. Discussion of this method, its characteristics, capabilities and results (for polar ozone loss and other applications) will be the subject of future papers.

Dependence on the vortex edge criterion. (p. 10051/6ff, 10052/4ff, 10057/16ff, figure 5, tables 1+2) The point that the number "vortex average ozone loss" does depend on the time period and the vortex edge criterion was made earlier (e.g. Grooß et al., 2008). . .

We have added references to the *Grooß et al.* study in sections 3.2 and 4 along the lines suggested by the reviewer.

...Most methods for deriving chemical ozone loss are limited due to data availability. However with a model or a data set with the coverage in space and time as MLS, such comparison is possible, i.e. a comparison with each publication of the value derived for the corresponding time range and vortex edge definition (e.g. as in Grooß et al., 2008). This may require some diligent work for Table 1, and as it stands now the paper is already very good, and this is not a necessary addition. But this comparison may potentially contribute to the understanding of parts of the differences. However, it would be at least interesting to see the comparison with Kuttippurath in table 2 using equivalent latitude 65°N instead of sPV=$1.4\times 10^{-1} s^{-1}$.

We agree with the reviewer that such an analysis would be informative, but also concur that it would require a lot of effort. Indeed, in some cases the literature may not provide enough information on specific criteria used in past studies to enable a direct comparison with our technique. A better approach would be a more collaborative future paper involving multiple teams and approaches, using a small number of agreed criteria and tests.
In lieu of such an effort, we have repeated analyses using the Kuttippurath equivalent-latitude-based criteria rather than our sPV one, as suggested by the reviewer. However, we note that the EqL criterion is considered less suitable in as it is only really applicable in periods when the vortex area is not changing significantly. Even in such cases, 65° EqL is probably too low for the Arctic and too high for the Antarctic because the latter vortex is typically larger. That said (and noted in the text), the results of an MLS Match-based loss estimation using an EqL criterion are included in Table 2, and we have removed the final sentence in section 5, instead adding a new paragraph as follows:

> We note that Kuttippurath et al. used an equivalent latitude-based vortex edge criterion, rather than the sPV criterion used for our study. Although equivalent latitude is less suitable in cases where the vortex area changes significantly with time, in order to allow a more direct comparison we have included results from our "MLS-Match" approach using the Kuttippurath et al. 65° equivalent latitude criterion in Table 2. The differences between the two MLS Match-based

estimates are nearly all smaller than the differences between either and the Kuttippurath et al. results.

---

p. 10043/13. PSCs do not form from gaseous sulfate species but from sulfate aerosol. Also heterogeneous reactions not only take place on the surfaces of the particles but also in the bulk of the liquid particles.

We had intended "species" to specifically distinguish from the "gaseous nitric acid" but agree that the wording was ambiguous. We have changed this word to "aerosols", and corrected the discussion of reactions. The new text reads (additions in italics):

> ...at which Polar Stratospheric Clouds (PSCs) form from gaseous nitric acid, sulfate ~~species~~ *aerosols*, and water vapor [citations]. Chemical reactions on the surfaces of *and within* these cloud particles liberate atmospheric chlorine...

---

p. 10044/19. Here the paper by Müller et al. (2005) should also be mentioned that discusses the ideas of Michelsen et al. and Plumb et al.

We have added this reference, as well as Plumb (2007), which also expands upon the ideas of Michelsen et al. and Plumb et al.

---

p. 10045/6ff. You could mention the advantage of using limb sounding as MLS over solar occultation data (POAM, ILAS) due to much better statistics.

This point is made later on in the manuscript (p. 10048/6ff), and we didn't feel that this historical discussion was a suitable additional place at which to preview it.

p. 10045/9. As an example for determining chemical kinetic constants from atmospheric observations, I would rather propose to cite von Hobe et al. (2007) and/or Suminska-Ebersoldt et al. (2012) as the Schofield study has a rather large uncertainty (see e.g. fig 2-11 of the 2010 WMO ozone assessment)

We have added the two references suggested by the reviewer.

---

p. 10048/section 2.3. The matches with a strong divergence in sPV between the central and flanking trajectories are discarded similar to the ozone sonde Match studies by Rex et al. The idea behind this is of course to discard matches with a possibly large trajectory error. However, this implicitly means that also areas of large wind divergence are avoided systematically. Is it possible to add one point in the panels figure 2 that represents an evaluation without the Flank divergence criterion? Likely it should be in the vicinity of the 500 km point, but it would be better to show it in this figure.

We have added this point (noting it in the figure caption and text) and, as expected, the results are little different from the 500 km case (∼0.1 ppbv/hour less loss).

---

p. 10049/8ff. Good. It is important to check whether the destination observations is also within the polar vortex, which has not been done always in the case of the sonde Match (see e.g. Fig. 7 of Grooß et al., 2008, ACP)

Noted.

---

p. 10053/14f. How exactly are the loss rates integrated over the winter? Is it done on descending surfaces following the average vortex descent rate? If so, how is the descent rate determined? By the use of MLS N2O?

No, these estimates are simply accumulated along constant potential temperature surfaces. We have added a comment to that effect in the text in section 3.3. Specifically:

> ... Integrating these losses from 1 January to 1 April (Fig. 3f) *on constant potential temperature surfaces* gives peak loss of slightly larger than 1 ppmv at 450 K...

---

p. 10054/3f. Potentially other possible error sources could be mentioned as errors in the wind fields taken from the meteorological analyses or interpolation errors.

These may indeed be the numerical sources of such errors, but we were trying in this discussion to describe how such errors manifest in our ozone loss estimation. To make that clearer we have modified the text as follows (additions in italics):

> Periods and locations for which significantly non-zero N_2O rates of change are calculated are therefore indicative of errors in the transport calculations used in the Match approach*, for example due to errors in the wind fields and/or biases introduced by their interpolation to the trajectory locations.* The two main routes whereby *such* transport errors can affect inferred ozone loss rates are inaccuracies in the trajectory calculations' depiction of diabatic descent within the polar vortex and of mixing across the polar vortex edge.

---

p. 10058/4. Mention also the chosen vortex edge criterion.

Done (in conjunction with adding the Grooß et al. reference).

3 Response to reviewer #3

The authors have applied Match technique approach to quntify the polar stratospheric ozone loss using the product of MLS Lagrangian Trajectory diagnostics and MLS measurements. They have also investigated several uncertanties on the estimation of ozone loss using different crtieria in this method. Then compared the current work with previous studies for Arctic winter 2004/05. Obviously this work has lower Arcitc ozone loss compared with most of other published works. For the Antarctic winter, this study also underestimates the partial column ozone loss (350 – 880 K) compared with Kuttipurath et al. Although the authors discussed this in Page 10058, but I think it would be better to explain more why this method produces lower ozone loss than others. The authors also estimates Arctic and Antarctic ozone loss for most of MLS period (2004 – 2013).
The paper is well organised and written and the objective of the paper is quite clear. This work extends the quantifying the ozone loss for Arctic and Antarctic winter and will be definitely used for the next WMO ozone assements and has a wider interest to the scientific community for studying the polar stratospheric ozone loss. The paper is suitable for publication at ACP, here I just have a few minor comments.

We thank the reviewer for their nice assessment of the paper. We agree that investigation of the reasons why this study gives smaller ozone loss estimates than previous quantifications would be valuable. However, we feel that such a study is beyond the scope of this paper. To be performed properly, such a study needs the active involvement of scientists leading the other quantification efforts because, as in this manuscript, the sensitivities of those studies to their inherent assumptions, and to criteria such as the definition of the vortex edge, need to be considered. Such studies have been performed in the past (e.g., the *Harris et al.* reference in the manuscript), and it may indeed be time to repeat and update such an exercise.

1) It is still vital to calulate ozone loss based on the information from the products of MLS LTDs. It seems that the products used from LTDs include (trajectory lati- tude,longitude, time, potential temperature and temperature, sPV, equivalent latitude (EqL)) in Page 10047 Lines 22-25. The current work is mainly based on using sPV. Can you recalculate the ozone loss using EL which have been also widely used by other researchers to see if there is any large difference?

This was also suggested by reviewer #1, and has been performed. The results of the loss quantification using an EqL-based vortex edge criterion are included in Table 2, and the associated discussion has been modified by deleting the final sentence in section 5 and adding the following new paragraph:

> We note that Kuttippurath et al. used an equivalent latitude-based vortex edge criterion, rather than the sPV criterion used for our study. Although equivalent latitude is less suitable in cases where the vortex area changes significantly with time, in order to allow a more direct comparison we have included results from our "MLS-Match" approach using the Kuttippurath et al. 65° equivalent latitude criterion in Table 2. The differences between the two MLS Match-based estimates are nearly all smaller than the differences between either and the Kuttippurath et al. results.

---

2) Fig2c shows the mean sPV for all points along the match trajectoies as a function of mean EL, why there is no EqL value larger than 75N (I know the MLS is only observed below 82N)?

Figure 2c shows estimated vortex average ozone loss versus the *mean EqL of all the tracked airmasses within the vortex*. These airmasses cover the full range of EqL from the vortex edge to 90°N. Choosing more stringent vortex edge criteria skews towards higher equivalent latitudes, increasing the mean EqL as would be expected.

3) Fig3(d). Why the ozone hour rate of change around 450 K is always positive from 1 Jan to mid-Jan 2015?

This probably reflects transport errors (e.g., underestimation of the impact of descent, which brings ozone-rich air from higher in the vortex, or inaccurate estimation of the impact of mixing across the vortex edge). These issues are discussed in more detail in section 3.4 of the manuscript.

---

4) delete "L." in Line 6 Page 1044.

The "L." is there to distinguish this paper from the W. Feng paper from the same year.

---

5) Line 2 Page 10045, change "sondes" to "ozonesondes"

Changed here and elsewhere throughout.

---

6) Lines 17, 27 in Page 10047, is it "analysis" or "reanalysis"?

"Reanalysis" is a specific term used for long term runs that are performed with a fixed assimilation system and underlying GCM (e.g., MERRA, ERA-Interim). Neither GEOS-5.1 or GEOS-5.2 are "reanalyses", they are "analyses".

---

7) Line 8 Page 10049, for the sPV, do you used the samer criterion for all altitude levels?

We do, as this is generally a robust definition of the vortex edge over a large vertical range. We acknowledge that other values may be more suitable, on occasion, at higher altitudes but, for simplicity, have opted to retain a single value here. We have added a new discussion of this point in section 3.1 as follows:

> The scaling of potential vorticity enables the same value to be used as a vortex edge criterion throughout the vertical range. An additional advantage of sPV-based rather than equivalent latitude-based vortex edge definitions is that the same value is a good measure of the vortex edges in both hemispheres and throughout most of their lifecycles. Sensitivity of our results to all these criteria is quantified in Sect. 3.2 while Sect. 4 includes discussion of loss estimates using an equivalent latitude-based vortex edge criterion.

---

8) Line 10054, can you explain more about "decent assumption" and "mixing assumption"?

We have revised the wording to, we hope, make this clearer. The new text is (additions in italics):

> The accuracy of MLS Match-based ozone loss estimates is further quantified by applying the same calculations to the MLS observations of N_2O, a long-lived tracer whose chemical rate of change is negligible on the timescales considered here. Periods and locations for which significantly non-zero N_2O rates of change are calculated are therefore indicative of errors in the transport calculations used in the Match approach*, for example due to errors in the wind fields and/or biases introduced by their interpolation to the trajectory locations*. The two main routes whereby *such* transport errors can affect inferred ozone loss rates are inaccuracies in the trajectory calculations' depiction of diabatic descent within the polar

> vortex and of mixing across the polar vortex edge. Inaccurate quantification of both of these processes, along with other transport errors (such as potentially erroneous identification of the matches themselves), will all contribute to errors in the ozone loss estimates. However, disentangling all these contributions is not feasible (at least not when only one long-lived trace gas, such as N_2O, is considered). Accordingly, for simplicity we ~~estimate~~ *compute two estimates of* the potential contributions of such transport errors to our ozone loss ~~estimates by ascribing~~ *calculations by assuming that* the observed N_2O changes *are* exclusively *due* to errors either in descent or in mixing, ~~and~~ *calculating the magnitude of the transport errors required to explain the observed* N_2O *behavior in each case, and then inferring their corresponding impact on ozone loss estimates. We* refer to these two ~~cases~~ *error estimates* as the "descent assumption" and the "mixing assumption" hereinafter.

---

9) Line 1 Page 10061 why "temperature"?

Temperature is needed to estimate density for a given pressure, an essential part of the column calculation. We have revised the text accordingly (additions in italics):

> Table 2 compares Arctic column ozone loss estimates from our approach (computed from the mixing ratio losses given in Fig. 6 using the mean vortex-average GEOS-5 temperature profile for each winter period *to estimate density for a given pressure level*) to those reported by...

---

10) Table 1, remove "W." or "W. Feng et al. (2007)"

Again, the W. was inserted (automatically) to distinguish from the L. Feng paper of the same year.

11) Table 2, change “MLS Match” to “This work”, Also use “Kuppippurath et al.”, it would be better also include published year.

We made the first change. However, as the Kuttippurath publication dates are detailed in the caption, adding them to the row labels would be superfluous.

12) Figure 2a, why uses “1000s”? The number in the Figure sometimes is overlapped, need to replot it to make it clear.

We have changed “1000s” to “thousands” to be clearer. We tried various approaches to “tidying up” the annotations. However, short of omitting them, we were unable to find a clean approach. We take the view that the annotations are useful in regions when they are not too cluttered and, when they are cluttered, there is arguably less need to distinguish the points anyway.

4 Summing up

We believe that our responses above and the revised manuscript sufficiently address the helpful and thoughtful comments and concerns of the reviewers.