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Interactive comment on "Delayed Recovery of mid-latitude lower stratospheric Halogen Loading" *by* Andreas Engel et al.

Anonymous Referee #1

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Review of Delayed Recovery of mid-latitude lower stratospheric Halogen Loading By Engel, Bönich and Ostermöller

The authors lay a foundation for a new approach to computing EESC that attempts to account for some issues with this parameter. In addition, they draw the conclusion that recovery of mid-latitude lower stratospheric halogens (and ozone) will be delayed by 10 years compared with current projections.

I have many issues with this work. These start with the idea that any formulation of EESC is a certain metric for 'recovery'. It depends of course upon projections for many constituents, and therefore depends on their lifetimes and various other uncertainties. It gives a reasonable estimate for recovery, and a useful tool for quantifying the sensitivity of ozone to ODSs along with all of the other factors that influence ozone (solar,

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volcanoes, qbo, etc.). I am generally skeptical that a change of a few percent in EESC changes anything about our understanding of recovery, or will make it easier to discern 'recovery' as time goes on. My concerns include some broad discussion points that I think can lead to misunderstanding as well as lack of referencing to a number of papers that are fundamental to the prior approaches to EESC calculation and use. There are also some overarching issues discussed below.

Overarching issues

1. There are different ways to think about the fractional release of ozone depleting substances. One way is to consider an age spectrum for elements, and to examine such factors as the maximum height achieved by parcels corresponding to each element. Statistical relationships can be obtained that make it possible to generalize the age spectrum to make sense for reactive gases. In older elements, most or all of the initial ODS has been destroyed. In younger elements, most or all of the initial ODS remains. The statistical relationship between the age and likelihood of destruction changes for source gases depending on their loss characteristics – specifically, how high must an element rise above the ozone peak in order to have experienced ODS destruction. Various papers take this approach – Hall (2000); Schoeberl et al. (2000); Schauffler et al. (2003); Schoeberl et al. (2005); Douglass et al. (2008). Distributions of various trace gases with a range of lifetimes show the validity of this approach as their lower stratospheric distributions carry information about the age spectrum. Waugh et al. (2007) show that the relationship of inorganic chlorine to mean age varies in different model implementations of the same meteorological fields, emphasizing the importance of a simulation reproducing not just the lower stratospheric mean age distribution but also distributions of various long-lived gases to attest to the realism of the age spectrum. It is especially important that appropriate relationships between maximum altitude and age are reproduced for all elements of the age spectrum. When this is achieved for a number of gases with varying lifetimes, then the relationship of fractional release as obtained from mean age, an observed gas, and the entry value to the stratosphere

estimated using the mean age makes sense as long as the dependence of the entry values is not a strong function of time.

These ideas become problematic if the sources are time dependent (and in particular when the sense of the time dependence changes from increasing to decreasing) especially when attempting to derive information from compact relationships that are obtained from observations for ODSs with a wide range of lifetimes. It may be necessary to account for the different values for entry to the stratosphere for various elements of the age spectrum rather than using the mean age to estimate that average. None of these things are well determined by observations, hence the reliance on some estimated width of the age spectrum. Many of these issues are less important in the current era, especially for the gases with lifetimes as long as CFCl3 and CF2Cl2, now that production is curtailed and the flux into the atmosphere from 'banks' is small compared to the present atmospheric burden.

The ideas presented above, where the contribution of each element in the age spectrum to an observed mixing ratio is controlled by its entry value, its path and the specifics of the loss for each ODS, are equivalent to the idea of 'arrival times' that vary for each ODS. Arrival times replaces one set of unknowns for another – by weighting the age spectrum with loss, you get a new 'time' quantity, different for each species, that controls the amount of a substance that you measure. This is a different twist on the same information that says that the long-lived elements control the amount that is destroyed, and applies a weighting function (different for each ODS) that is based on maximum altitude and probability of destruction to the same age spectrum . For a long-lived gas like CF2Cl2 more elements of the age spectrum contribute to the observed amount than for a shorter-lived element like CFCl3, thus more elements are in play in the distribution of arrival times. (Conversely, for a longer-lived gas fewer elements contribute to the observed inorganic chlorine etc.) From a physical point of view, I don't see the advantage of saying that the mean arrival time, shorter than the mean age, varies with species and that the age spectrum is 'inappropriate for a reactive gas' than

saying that the age spectrum is a statistical property of the flow, and that the amount of the gas observed is controlled by the statistical properties of the elements that produce the age spectrum and their ODS values at the time of entry to the stratosphere.

I think that to be convincing on the subject, the paper would have to explain this formulation in the context of work by the authors cited above and others published during the 2000s rather than relying on Plumb et al. 1999 which was primarily an attempt to generalize correlation relationships that depended on the time dependence of the source gases during a time when CH3CCl3 (methyl chloroform) was decreasing rapidly due to stoppage of production and its short life time.

2. The assumption underlying the projection on 'recovery' – that stratospheric dynamics and chemistry remain unchanged – is certainly false. The stratosphere is cooling and virtually all models project a speedup in the Brewer Dobson circulation. Although there is no consensus on details of how the circulation will change in the extratropics, and uncertainties in circulation change drive the differences in projections of the ozone layer for 2100, it is not sensible to make a projection that could only be true if the changes that are already observed to be occurring did not take place. The paper title asserting 'delayed recovery' is therefore misleading and unnecessarily alarming given the observational evidence that the column ozone loss at middle latitudes has been effectively limited ($\sim$4%, WMO 2010; WMO 2014) by the Montreal Protocol and its amendments.

3. The overall significance attached to EESC is unreasonably large. As the authors discuss, EESC is commonly used to separate ozone sensitivity to ODS build-up from other factors. The projection of delayed recovery comes from a lower value for EESC in 1980, thus return to 1980 will occur later. Does the new formulation change the ozone sensitivity that is obtained from the data record? Is there improvement in the statistical significance of the ozone sensitivity to chlorine change (e.g., is the overall fit to the prior record improved). Is there any observational evidence that shows that this formulation is both correct and beneficial? Does this formulation agree substan-

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tially better with results from a 3D simulation (that passes the CCMVal transport tests) where the EESC can be calculated directly, where the mean age can be calculated either from pulsed experiments and these ideas can be fully tested, including physical importance? Finally, what would this mean with respect to the non-volcanic ozone loss already accrued by 1980 discussed by Shepherd et al. (2014)? Also note that Shepherd et al. compare the 'old' EESC with their calculation (summing up contributions to inorganic chlorine and bromine) and demonstrate that they agree.

Specific Major Comments

P 2 Line 5 Importance of EESC uses is overstated e.g., Tilmes et al. 2009 do not use EESC to discuss geoengineering. They compare their result to Newman et al. 2006, limited to polar ozone. Shepherd work shows that the conventionally derived EESC (Newman et al. 2007) agrees with the 3 D model result for the sum of inorganic chlorine and 60 times inorganic bromine . . . Weatherhead and Anderson (2006) use EESC as it is intended, as a parameter that generally describes the evolution of ozone depleting substances, but the subject of the paper is detectability of ozone trends and the discussion of the many factors that affect ozone, short term trends, and identification of 'recovery'.

An important question left undiscussed is whether and how this reformulation would change any of these results.

Page 3 Line 18 – EESC is 'influenced by . . . fractional release factors' This suggests that these factors are somehow invariant properties of the gases, whereas they are inherently tied to the meteorological factors that control the overturning circulation and mixing.

Page 4 and elsewhere: we discuss the interaction of chemistry and transport Chemistry and transport are not actually 'interacting' for these long-lived gases. The loss is a function of parcel path. The loss does not affect the parcel path, and therefore does not affect the transport. So what you are discussing is loss along a parcel path rather

than an interaction.

Page 4 section 2 Generally speaking, most of the loss takes place in the tropics (e.g., Prather et al., JGR 2015) N2O 81% between 24 and 40 km; 76% between 30S and 30N.

This section goes through a lot of details but the basic picture is not changed. A) the relationship of mean age, age spectrum, and the fractional release is not simple, as the contribution of each element to destruction of the ODS depends on parcel path. B) when comparing various species to their 'entry' values, in an ideal situation you have data on species of varying lifetimes so that you can obtain information about the mean age and the age spectrum from their differences; C) if the circulation is altered in any way such that the statistical relationships between age and path are altered for various elements of the age spectrum, then the empirical relationships will change. In fact, measurements of an inert gas such as SF6 in combination with measurements of various ODSs would provide information as to whether or not the age spectrum was changing.

Page 8 – the arrival time distribution and the release time distribution are 'closely linked'. This seems like it took a lot of work to get to a statement that follows directly from conservation. Also and probably more important – you don't ever cut to the chase for the simple physical principle. Lastly – the 'path independent fractional release' for a given mean age discussed in section 2 must have the relationship between loss, altitude and path imbedded in it – without some elements in the age spectrum reaching high altitude there will not be that much loss (at least for gases like CF2Cl2).

Section 4 In the end, you reach something that could be a simply stated conclusion. When EESC is growing rapidly, the relationship of mean age to older elements of the age spectrum is complicated because an entry value that is a simple function of the mean age does not take into account the very low entry values for the oldest elements of the spectrum. When EESC is decreasing, the reverse situation becomes the norm –

the old elements of the age spectrum have higher entry values than might be inferred from the mean age.

This work does not make a strong case for using the new formulation of age spectrum to project ozone recovery, rather it makes a case for uncertainty of EESC (although the values in Table 3 differ by less than 5% for mean age of 3 years), as well as a case for understanding the limitations of EESC. Given that midlatitude ozone loss is only $\sim$ 4%, it seems unlikely that the data up till now will differentiate one formulation from the other. It also seems unlikely that a difference in return to 1980 values would be discernable given all the other sources of variability (e.g., Mahieu et al., Nature, 2014) – the (difference between max and 1980)new – (difference between max and 1980)ccurrent is less than 10%.

Other Comments

Abstract: why 'can lead'? why not 'lead'?

Throughout: Please be careful and precise with 'depletion' and 'loss'. Loss is often natural. Depletion means loss greater than natural loss.

Conclusions: We suggest that this new method to calculate EESC be used to estimate the time of recovery of inorganic halogen to 1980 values – I suggest that you don't use EESC to estimate the time of recovery except in the broadest sense. Among other reasons – we don't have enough measurements of inorganic halogens in the 1980s to be confident of the starting value.

You propose comparing inorganic halogen levels from a full model calculation with the new EESC – I don't understand why you have not already used full model calculations (easily available through CCMI) to make this comparison.

Finally – if the new EESC formulation were shown to make a difference in the analysis of ozone time series, then that would be an important reason for its use. As it stands, this paper attempts to address a particular limitation to the current formulation for the

relationship of mean age, tracers, the age spectrum and fractional release. The discussion of the formalism does not make clear the reason for doing it clear from a physical point of view, and the geophysical significance of the result is overstated.

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