

## Answer to Reviewers

We would like to thank all four reviewers for taking the time to go through the manuscript and giving us valuable feedback, which will help in improving the manuscript. Part of the criticism mainly from reviewer #1 and #2 is on the general use and concept of EESC and its use for the projection of a recovery date. This may be partly due to the choice of the title, which suggests a real change in recovery date. Actually the paper is mainly about the general concept of deriving EESC and therefore the title is indeed misleading. We suggest to change the title to “A refined method of calculating EESC” to emphasize that the idea is mainly to develop a concept which is mathematically consistent and physically more meaningful than the present formulation. We also understand the many limitations of the concept of EESC and that EESC will always be only a proxy for inorganic chlorine. Yet, EESC has some values, as it is a good proxy for one single parameter which has been subject to change due to a direct anthropogenic impact, namely the emission of halocarbons. It is therefore of considerable interest to understand, in particular in the context of the Montreal Protocol, when this impact is reversed and when the anthropogenic influence on this parameter will have reached a value which is deemed sufficiently close to background conditions. While the Brewer-Dobson circulation is expected to change in the future with considerable impact on inorganic chlorine in the stratosphere, it is exactly the idea behind EESC to have a measure of changes in this sole parameter of EESC. We also show in our revised manuscript that our new formulation is a reasonable proxy of inorganic halogen loading for the future on a given pressure level. We have added a new section (4.3.) to the manuscript which (i) compares our new formulation to that currently used based on a model calculation with fixed dynamics and (ii) shows that even under a changing circulation EESC is a reasonable proxy for inorganic halogen loading. Due to these additional comparisons with model calculations, we have added three additional co-authors to the paper, who provided model data for comparisons. We have also changed the abstract to reflect the changes. It does not contain a specific return data anymore but only mentions the time shift compared to the current formulation and also discusses the results from the model comparisons. New abstract:

***Abstract.** Chlorine and bromine atoms lead to catalytic depletion of ozone in the stratosphere. Therefore the use and production of ozone depleting-substances (ODS) containing chlorine and bromine is regulated by the Montreal Protocol to protect the ozone layer. Equivalent Effective Stratospheric Chlorine (EESC) has been adopted as an appropriate metric to describe the combined effects of chlorine and bromine released from halocarbons on stratospheric ozone. Here we revisit the concept of calculating EESC. We derive a refined formulation of EESC based on an advanced concept of ODS propagation into the stratosphere and reactive halogen release. A new transit time distribution is introduced in which the age spectrum for an inert tracer is weighted with the release function for inorganic halogen from the source gases. This distribution is termed the “release time distribution”. We show that a much better agreement to inorganic halogen loading from the chemistry transport model TOMCAT is achieved than using the current formulation. The refined formulation shows EESC levels in the year 1980 for the mid latitude lower stratosphere, which are significantly lower than previously calculated. 1980 marks the year commonly used as benchmark to which EESC must return in order to reach significant progress towards halogen and*

*ozone recovery. Assuming that – under otherwise unchanged conditions - the EESC value must return to the same level in order for ozone to fully recover, we show that it will take more than 10 years longer than estimated in this region of the stratosphere with the current method for calculation of EESC. We also present a range of sensitivity studies to investigate the effect of changes and uncertainties in the fractional release factors and in the assumptions on the shape of the release time distributions. We further discuss the value of EESC as a proxy for future evolution of inorganic halogen loading under changing atmospheric dynamics using simulations from the EMAC model. We show that while the expected changes in stratospheric transport lead to significant differences between EESC and modelled inorganic halogen loading at constant mean age, EESC is a reasonable proxy for modelled inorganic halogen on a constant pressure level.*

10 One major fundamental comment to the paper, which was raised by reviewers 1 and 2 was that chemical breakdown of trace gases is not influenced by the transit time in the stratosphere, but that the location of transport pathway, and in particular the maximum path height (MPH) is the fundamental parameter, as discussed e.g. by (Hall, 2000) . This is in principle correct and we had partly addressed this in our paper, but probably not sufficiently. We had stated in the paper:

*“Chemical loss is not uniform throughout the stratosphere, as it depends in most cases on the actinic flux at short wavelength.*

15 *In the seasonal mean, the chemical lifetime will in general decrease with altitude and increase with increasing latitude. The chemical loss is thus very inhomogeneous, but on average, it is expected that the longer a fluid element remains in the stratosphere, the larger the integrated chemical loss will be. Also, as the loss for most species with photochemical sinks mainly occurs at higher altitudes, it is expected that, on average, longer transit times will be associated with shorter lifetimes.”*

Even better than the MPH, as stated in previous papers (Schoeberl et al., 2005;Hall, 2000), the total UV exposure along a transport path is the best measure for photochemical loss. This photochemical exposure as function of transit time is not available and will differ for different transit pathways even if they may have the same transit time. Our approach relies on the assumption that **on average** there is a relationship between transit time and photochemical loss, so that on average the longer an air parcel has been in the stratosphere, the larger the chemical loss and thus the fractional release will be. Therefore, fractional release can be related to transit time. Figures 4, 6 and 8 in (Hall, 2000) actually show joint distributions showing that on average there is a correlation between transit time and MPH. This relationship between photochemical loss (for which MPH is also only a proxy) and transit times is actually the basis of the work of (Plumb et al., 1999) and (Vohralik et al., 1998) on which we base our concept and the parameterization of chemical loss. They were able to show that they could detrend correlations between different trace gases using the parameter of mean arrival time based on the concept that there is a clear relation between mean age and the photochemical degradation weighted mean arrival time. They state that “molecules arriving at X with long arrival times will, on average, have spent more time exposed to chemical loss and will have sampled atmospheric regions where photochemical loss is greater”. This is exactly the prerequisite needed for our approach to describe the chemical loss as a function of transit time only without adding the path as an additional parameter. A detailed reply and how we addressed this concern is given in the individual reply to the reviewers (see foremost point 1 of reviewer 1).

In the following we will now answer the points raised by the reviewers point to point. As there are four reviews, we will partly refer to answers given to other reviewers. Our answers are shown in italic and changes to manuscript are shown in red.

## Reviewer 1

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

5 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, 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.

*The reviewer is correct in pointing out that some reference to previous work was missing. We apologize for this and have now included these papers in the discussion (see details given below). We have further changed the title of the manuscript to “A refined method of calculating EESC” in order to lay the emphasis more on the principal method rather than on the issue of recovery date. In this respect we would also like to point out that EESC is a parameter describing the evolution of one single parameter (inorganic halogen loading), which has been subject to change due to a direct anthropogenic impact in the past, namely the emission of halocarbons. It is therefore of considerable interest to understand, in particular in the context of the Montreal Protocol, when this impact is reversed and when the anthropogenic influence on this parameter will have reached a value which is deemed sufficiently close to background conditions. We have included the following statement on EESC at the end of the first paragraph of the introduction:*

*Note that EESC is only a valid proxy for anthropogenic ozone depletion if all other parameters, especially atmospheric transport, are unchanged. A projection of a return of EESC to some specific level therefore does not imply that ozone will return to the same levels. EESC should thus be regarded as proxy for the impact of halogenated source gases on the ozone layer due to both anthropogenic and natural emissions. The recovery of the ozone layer is affected by other parameters in addition, especially changes in transport. EESC is therefore not a proxy for ozone recovery, but a proxy for the impact due to one single parameter, the halogen loading. In section 4.3. we discuss the validity of EESC as a proxy for inorganic halogen loading under the influence of changing stratospheric dynamics.*

35 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.

- 5 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.
- 10 Many of these issues are less important in the current era, especially for the gases with lifetimes as long as CFC13 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 longlived gas like CF2Cl2 more elements of the age spectrum contribute to the observed amount than for a shorter-lived element like CFC13, 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.

*First we would like to point out that the maximum path height MPH is also only a proxy, as what is of interest is the cumulative loss along a transit path, or as in our formulation for a transit time. The relation between transit time and chemical loss must only hold on average. There is no doubt that there will be parcels with long transit times but little loss. Nevertheless, on the average parcels with longer transit times do experience more loss, as also shown by e.g. (Plumb et al., 1999) on which a large part of work resides. This is also clearly visible in the work of (Hall, 2000) and the same approach has been used previously (Schoeberl et al., 2005; Schoeberl et al., 2000). This is the main point of criticism which we have addressed above. We think that there is sufficient evidence also from the tightness of correlations between mean age and chemical active species, that **on average** there is a correlation between the transit time and chemical loss, which is the prerequisite for our approach to describe fractional release as a function of transit time only. Note that we do not need to make any assumptions on the exact shape of this relationship, as this is included in the release time distribution  $G^\#$ . In order to give credit to other work on this issue and to emphasize this point, we have changed the manuscript in section 2 as follows:*

*The fractional chemical loss can be expressed in a very generalized way as  $(1-f(t^\wedge,p))$ , where  $f(t^\wedge,p)$  is a fractional release function, which is specific for each trace gas and will depend on the time the air parcel has spent in the stratosphere  $t^\wedge$  and on the path  $p$  it has taken during the transport, especially also of the maximum path height (MPH) (Hall, 2000). While the path or the MPH are not known, it has been shown that “molecules arriving at  $X$  with long arrival times will, on average, have spent more time exposed to chemical loss and will have sampled atmospheric regions where photochemical loss is*

greater” (Plumb et al., 1999). We therefore follow the approach that the fractional loss can on average be described as function of the transit time only. That chemical loss and transit time are on the average related to each other is also reflected in the tight observed correlations between mean age and tracer mixing ratios (e.g. Volk et al., 1997; Engel et al., 2002). While there will be fluid elements with very different paths and different chemical loss which have the same transit time the loss can on average be sufficiently well described as a function of the transit time. We therefore treat  $f(t^{\wedge},p)$  as  $f(t^{\wedge})$  only. This is also in line with the findings of Schoeberl et al. (2000), who showed that using an “average path approximation” with a “single-path photochemistry” and thus with a unique relationship between loss and transit time, global tracer-tracer correlations can be explained. This concept that loss can be described only as a function of transit time without considering the different transit pathways was also adopted by Schoeberl et al. (2005) in the derivation of age spectra. We use a mean age of 3 years for mid latitudes and of 5.5. years for high latitudes. Indeed, the path distribution for an air parcel with mean age of 3 years in the tropics, in mid latitudes and in polar regions is expected to show more variability than for air parcels investigated under similar conditions (e.g. latitude regions). As this analysis is restricted to one latitude band for one mean age level, we therefore approximate loss as a function of transit time only.

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 (WMO 2010; WMO 2014) by the Montreal Protocol and its amendments.

First of all, we have removed the term ‘recovery’ from the title of the manuscript, as it was indeed misleading. We have also emphasized in the introduction that EESC is a proxy for inorganic halogen loading only and not for the ozone layer and only under the assumption of otherwise unchanged conditions. We have also added a new section 4.3. in which we compare our new calculation with the current formulation and model results, showing that it gives much better agreement and also comparing EESC to future inorganic halogen loading in a chemistry climate model with changing stratospheric transport. We have added the following in the introduction:

Note that EESC is only a valid proxy for anthropogenic ozone depletion if all other parameters, especially atmospheric transport, are unchanged. A projection of a return of EESC to some specific level therefore does not imply that ozone will return to the same levels. EESC should thus be regarded as proxy for the impact of halogenated source gases on the ozone layer due to both anthropogenic and natural emissions. The recovery of the ozone layer is affected by other parameters in addition, especially changes in transport. EESC is therefore not a proxy for ozone recovery, but a proxy for the impact due to one single parameter, the halogen loading. In section 4.3. we discuss the validity of EESC as a proxy for inorganic halogen loading under the influence of changing stratospheric dynamics.

We have further added the section 4.3., here in particular the section comparing EESC to inorganic halogen in a full chemistry climate model. A reference to the sensitivity studies and model comparisons in section 4 has also been added at the end of the introduction:

We also present sensitivity studies of the new EESC formulation to different parameters and compare the different formulations of EESC to simulations of inorganic halogen loading from two different comprehensive three-dimensional atmospheric chemistry models.

## Comparison to model calculations with varying dynamics

*Under changing stratospheric dynamics (e.g. Butchart, 2014), it is expected that fractional release factors at a given mean age level will change (Douglass et al., 2008; Li et al., 2012; Ostermüller et al., 2017). Therefore, the inorganic halogen loading as a function of mean age would be expected to change even if all source gases remained constant in time. Under such conditions, EESC is not expected to follow ESC on a given mean age level. To estimate the validity of EESC as a proxy for inorganic halogen loading of the stratosphere, we have compared our new formulation to a free running chemistry climate model simulation. We used data from the EMAC model simulation RC2-base-04 from the ESCiMo project for this (Jöckel et al., 2016). This simulation covers the 1950–2100 time frame with simulated sea surface temperatures and sea ice contents. As described above, we again calculated fractional release factors from the model in order to have results which are internally consistent. Northern Hemisphere fractional release factors for the year 2000 are in good agreement with observation based fractional release factors (Newman et al., 2007; Laube et al., 2013) and therefore we used northern hemispheric data for this comparison. In addition to comparing ESC on a fixed mean age level we also compared ESC on a fixed pressure level to our new formulation of EESC. A similar comparison has been presented in Shepherd et al. (2014), who compared model ESC on a fixed pressure level to EESC on fixed mean age level (using the formulation of Newman et al. (2007)), showing good agreement. Figure 9 compares the time evolution of EESC for 3 years of mean age with model ESC at the 60 hPa level (corresponding to 3 years of mean age in the year 2000) and model ESC at 3 years of mean age. The year of 2000 and the corresponding level of 60 hPa was chosen, as we also evaluated fractional release factors in the year 2000 of the model run. As expected, ESC at 3 years of mean age deviates systematically from EESC, especially in the future when fractional release evaluated on a mean age surface changes significantly in the model. The agreement with ESC on a fixed pressure level is however much better. In this comparison EESC at 3 years of mean age would slightly overestimate ESC on a pressure level in the future and significantly underestimate ESC on a mean age level. The exact magnitude of changes in stratospheric dynamics is highly uncertain and also we expect ozone to follow a pressure surface rather than a mean age surface in the future. We therefore conclude that EESC is a reasonable proxy for the effect of halogen loading on stratospheric ozone, given the overall high uncertainties associated to the future evolution of stratospheric dynamics.*

*Further we have added in the discussion section:*

*We have shown that the long-term evolution of effective stratospheric chlorine (ESC, i.e. inorganic chlorine and bromine, the latter weighted in a similar way as in EESC to reflect the higher efficiency of bromine to ozone depletion) in the model deviates substantially from our calculation of EESC in a long-term model calculation with varying dynamics. However, we have also shown that the new formulation of EESC is a reasonable proxy for the evolution of inorganic halogen loading on a given pressure level. We therefore conclude that EESC is a reasonable proxy for future halogen impact on ozone.*

...

*As current climate models consistently predict an acceleration in the Brewer-Dobson circulation (Butchart, 2014), this will have an impact on the temporal evolution of inorganic halogen loading of the stratosphere. These expected changes in the Brewer-Dobson circulation would result in an earlier recovery of ozone at mid- and high latitudes (Eyring et al., 2010). These*

*changes are not included in the concept of EESC. However, we have shown that EESC is a reasonable proxy for ESC when ESC is evaluated at constant pressure level.*

- 5 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 substantially 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.

10 *In the model inorganic halogen (ESC, Equivalent stratospheric chlorine) can be directly determined from Cly and Bry. Even if there were no trends at all in organic halogen source gases, ESC on a given mean age level would change in the model, as fractional release is expected to change on a given mean age level. Interestingly, this change in fractional release is less pronounced on pressure levels. (Shepherd et al., 2014) compared the evolution of ESC at 50 hPa with EESC at 3 and 5 years of mean age. As the mean age on a given pressure level changes, these are to a certain degree two different things which are compared with each other, showing surprisingly good agreement.*

20 *A meaningful comparison of ESC and EESC is far from trivial. We have compared our new and the old formulation of EESC to an EMAC simulation (Jöckel et al., 2016) and found that the ESC in the model deviates substantially from EESC (both formulations), mainly due to long term changes of frf on mean age levels. As in (Shepherd et al., 2014), we find a much better agreement when comparing EESC on a mean age level with ESC on a given pressure level. On the other side, the mean age on the pressure level changes, so this comparison is not really valid. A more meaningful comparison between ESC and EESC can be done using a model run with fixed dynamics and changing ODS levels. We now show a comparison between our new formulation and a model run from the TOMCAT model (Chipperfield et al., 2017), which uses fixed dynamics for the year 1980. In this model run, a meaningful comparison can be performed and we show that there is much better agreement between our new formulation and the model calculation when compared to the old formulation. We have added a new section to the manuscript (section 4.3), called "Comparison of EESC formulations with model calculations of inorganic halogen loading" in which we show the comparison with the EMAC model (with changing dynamics) and the TOMCAT model (with fixed dynamics). Section 4.3. on comparison with fixed dynamics calculations:*

#### ***4.3. Comparison of EESC formulations with model calculations of inorganic halogen loading.***

40 *In order to evaluate our new formulation of EESC we have compared the results of our calculations with the inorganic halogen loading calculated from two comprehensive three-dimensional atmospheric chemistry models. Due to expected long-term changes in mean age on a given pressure level associated with the simulated changes in the Brewer-Dobson circulation (e.g. Butchart, 2014; Austin and Li, 2006), changes in fractional release factors on mean age levels are also observed in free running model calculations (Douglass et al., 2008; Li et al., 2012). We have therefore compared our new formulation of EESC to model calculations with changing and with annually repeating ('fixed') dynamics. To compare the new formulation with the formulation by (Newman et al., 2007), we used a model simulation from the TOMCAT model (Chipperfield et al., 2017), which was driven by a repeated meteorology, in this case for the year 1980. Effects due to changing dynamics, which are not included*

in the concept of EESC, will thus not impact this calculation, making it an ideal test bed for comparison of the two formulations. For long term changes, we have used model results from the EMAC model (Jöckel et al., 2016), which includes expected changes in stratospheric transport. As in general the relationship between mean age and  $Cl_y$  is very different for different models (Waugh et al., 2007), a direct comparison between EESC and ESC (Equivalent stratospheric Chlorine, calculated from model  $Cl_y$  and  $Br_y$  using the same sensitivity parameters for bromine as with EESC) is not meaningful, as differences may be due to different fractional release factors between models and observations. Instead we have used fractional release values derived from the models for 3 years of mean age and used these for the calculation of EESC using the formulations by Newman et al. (2007) and from this work. Fractional release factors were calculated from the model data using the methods of Newman et al. (2007) and Ostermüller et al. (2017) for this work. The fractional release factors were calculated for the year 2000, in order to be consistent with the observation based fractional release factors, which were derived mainly for the year 2000. To the EESC calculated in this way we added simulated inorganic chlorine and bromine at the tropical tropopause, which we propagated as an inert tracer. VSLs (very short lived substances) were treated in a similar way, using their tropical tropopause values as input, as loss in the troposphere cannot be neglected for these species. As no global stratospheric lifetimes are available for these species, it is not possible to apply the new formulation. Therefore, the VSLs were treated using the method of Newman et al. (2007). The differences are negligible, as the VSLs have rather slow long-term trends and both methods yield nearly identical results, as also discussed in Ostermüller et al. (2017). As some loss of  $CH_3Cl$  and  $CH_3Br$  occurs during the transport in the troposphere to the tropical tropopause, we have also used the time series of these two gases at the tropical tropopause rather than at the surface in these calculations.

### **Comparison to fixed dynamics model calculations**

For comparison of the two formulations to model calculations with fixed dynamics, we used a TOMCAT model run (Chipperfield et al., 2017), which is driven by repeated 1980 meteorology. This model run is available from 1960 through to 2016. The fractional release factors derived from the model for the northern hemisphere are significantly higher as function of mean age than the observed fractional release values. Southern hemispheric fractional release values for 3 years of mean age showed better agreement with observation derived fractional release factors (Newman et al., 2007). For this reason we compared simulated ESC from the southern hemisphere with EESC calculated using our new formulation and the formulation by (Newman et al., 2007), in both cases using fractional release values derived for the year 2000 model results.

Figure 8 shows the comparison between the modelled ESC and EESC for a mean age of 3 years calculated as described above, including all bromine and chlorine species included in the model and also including inorganic chlorine and bromine entering the stratosphere. As the differences between the two formulations of EESC are most pronounced for 3 years of mean age, we show this comparison for 3 years of mean age only. A much better agreement is observed when applying the new formulation than using the formulation by Newman et al. (2007), due to the improved treatment of the combined influence of transport and



*mixing on chemical loss. Remaining discrepancies between model ESC and EESC are most probably due to an imperfect parameterization of the loss time distribution  $G^{\#}$ .*

#### Specific Major Comments

- 5 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’.
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An important question left undiscussed is whether and how this reformulation would change any of these results.

- This is a good point. We do not think that the reformulation would affect the results of these studies in a qualitative way. It is only when EESC is used in a quantitative way, e.g. in the projection of possible changes using different scenarios and only when frf values are significantly lower than 1 that this reformulation is expected to have a significant impact. We have added the following sentence in the conclusions:*
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- As the suggested reformulation of EESC does not affect the principal behavior of the temporal evolution of EESC, we do not expect this reformulation to lead to substantial changes, which could impact the changes of studies using EESC except for those which have used EESC to project EESC recovery.*
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- 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.
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*This was certainly not the intention of that statement. But fractional release factors should be independent of tropospheric trends of ODS. In order to make this clearer we have added the following statement behind this sentence:*

- While all these factors may vary with time, e.g. due to changes in stratospheric circulation (Douglass et al., 2008;Li et al., 2012), especially fractional release factors should not depend on the tropospheric trends of the trace gases (Ostermüller et al., 2017).*
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- 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.
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- We only partly agree. Of course the chemistry has a feed-back on the transport via radiation and heating rates. Nevertheless we suggest to rephrase the title of this section to “On the influence of transport and tropospheric trends on chemical active species”.*
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- Page 4 section 2 Generally speaking, most of the loss takes place in the tropics (e.g., Prather et al., JGR 2015) N<sub>2</sub>O 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,
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measurements of an inert gas such as SF<sub>6</sub> in combination with measurements of various ODSs would provide information as to whether or not the age spectrum was changing.

5 *We are not sure, what the reviewer is suggesting here. In principle we agree to the statements, but we would like to emphasize again, that the current formulation of EESC does not take into account that fractional release is in any way dependent on the transport path or transit time. In our new formulation this dependency is added, by using a transit time dependent approach for fractional release (and assuming that on average longer transit times are related to more chemical loss). Regarding the possibility of deriving age spectra and changes of age spectra from observations, this is a very different subject and not the scope of this paper. And indeed, fractional release as a function of mean age is expected to change. This is, however, not*  
10 *included in EESC, neither in the old nor in our new formulation.*

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  
15 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 CF<sub>2</sub>Cl<sub>2</sub>).

*This touches again on the main point addressed at the top of our reply. We do not suggest that fractional release is path independent but rather we suggest that this path dependence can be treated as a transit time dependency given that, on average, longer transit times are associated with more time spent in the chemical loss region in the upper tropical stratosphere and thus more chemical loss.*  
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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.  
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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 ~ 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) current is less than 10%.  
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*In order to avoid a wrong impression that might have been caused by the title, we have changed the title, to emphasize that the main purpose of this paper is a new, mathematical consistent, formulation of EESC. In the same time, we have slightly different understanding of what EESC should provide, as its main purpose is to look at the halogen-induced impact on the stratosphere, where it can be used as proxy and also as metric to assess when this effect is reduced to levels which are comparable to those present in the stratosphere at some reference time set (rather arbitrarily) to 1980. We have made this clear by including the following statement in the introduction:*  
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*Note that EESC is only a valid proxy for anthropogenic ozone depletion if all other parameters, especially atmospheric transport, are unchanged. A projection of a return of EESC to some specific level therefore does not imply that ozone will return to the same levels. EESC should thus be regarded as proxy for the impact of halogenated source gases on the ozone layer due to both anthropogenic and natural emissions. The recovery of the ozone layer is affected by other parameters in addition, especially changes in transport. EESC is therefore not a proxy for ozone recovery, but a proxy for the impact due to one single parameter, the halogen loading. In section 4.3. we discuss the validity of EESC as a proxy for inorganic halogen loading under the influence of changing stratospheric dynamics.*  
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*We have further added the section 4.3. which describes the comparison of our new EESC formulation and the formulation by Newman to model calculations.*

#### Other Comments

- 5 Abstract: why ‘can lead’? why not ‘lead’? *Has been changed.*

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

- 10 *Thank you, we have made sure that depletion is only for anthropogenic and loss for natural and anthropogenic loss. In most cases loss is used with respect to ODS and not with respect to ozone.*

- 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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- We only partly agree to this statement that EESC should not be used to project a recovery. EESC has been used regularly in the WMO ozone assessment reports to estimate the level of recovery already achieved due to the regulations of the Montreal Protocol and also to project future halogen loading of the stratosphere and possible recovery dates for this halogen loading to the admittedly partly arbitrary benchmark level of 1980. EESC certainly has limitations, but on the other hand it has advantages over a model calculation. This is to one part that there are no changes in circulation considered (the extend of which are not well known) and that EESC is based to a very large degree on observations. Not only the tropospheric time series are observation based but also the chemical loss (fractional release factors). With respect to EESC projections not including changes in circulation makes it a valid tool to assess the rate of recovery of the parameter which is most easily influenced by anthropogenic activity and in end political action, namely the halogen loading. Of course, this changes is for “all other things being equal”. The suggested comparison with model calculation is far from trivial, as in these model calculation ESC (equivalent stratospheric chlorine, e.g. the sum of  $Cly + 60$  or  $65$  times  $Br_y$  from the model) is dependent on many more factors, especially the expected changes in stratospheric transport. These changes can only be disentangled in model calculations without long term changes in circulation but with changes in trace gases, e.g. calculations with fixed dynamics. We have performed such a comparison for a model with changing dynamics (EMAC) and a model with fixed dynamics (TOMCAT). The comparison is included in a new section 4.3. The change in dynamics in the EMAC model results in large differences between EESC and ESC when ESC is evaluated on a given mean age level, but reasonable agreement when ESC is evaluated on a fixed pressure level. The comparison with the TOMCAT model and the old formulation of EESC and our new formulation shows that a much better agreement is found using the new formulation. This is discussed in the new section 4.3. and the conclusions section, as explained in the answer to the major question #3.*
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#### 45 **Reviewer 2**

This paper describes a reformulation of the EESC metric that attempts to account for the difference in stratospheric mass transport through photochemical loss regions vs. transport outside of the loss regions. The result is a relatively older mean age used to calculate EESC and thus a delay in future mid-latitude lower stratospheric EESC values to 1980 levels of roughly one decade.

Unfortunately, the paper has a number of fundamental flaws that make it unsuitable for publication. The major issues include (1) the absence of path height dependence in the fractional release and modified age distributions, (2) the assumption of constant stratospheric dynamics and photochemistry over nearly a century for the results to be valid, (3) the relatively minor role of EESC in future mid-latitude ozone depletion compared to N<sub>2</sub>O and (4) the absence of many relevant references and discussion of previous work.

Main comments:

1. The authors mention in Section 2 that chemical loss is not uniform throughout the stratosphere and argue that on average the longer a fluid element remains in the stratosphere the larger the integrated chemical loss will be. This is not the case. The highly nonlinear loss dependence on altitude and latitude make the path an air parcel has taken much more important than the time an air parcel has resided in the stratosphere. Hall (2000) described the concept of maximum path height and the relationship between the path height and age distribution. Hall showed quite clearly that the mass fraction of an air parcel that has passed above some height, such as the height above which photochemical loss is rapid, is determined by the transport due to mass continuity and not the circulation, including mixing, that determines age distributions. Thus, the age distribution is only weakly linked to stratospheric photochemical destruction of any trace gas. This means that it is the circulation due to mass continuity that is most relevant in the estimation of EESC.

The circulation due to mass continuity is essentially the residual circulation and it has been shown, such as by Birner and Bonisch (2011) that the transit time due to the residual circulation is at most 3.5 years in the polar regions where the air parcels reached a minimum pressure of less than 0.1 hPa, well into the mesosphere, before descent into the polar vortices. This also implies that transit times throughout the mesosphere are actually less than 3.5 years. Mixing of air horizontally acts to increase the age of the stratosphere nearly everywhere (e.g. Garny et al., 2014) but not necessarily the maximum path height of a parcel and thus it's photochemical loss. The mixing is what drives the old tail in the age distribution and this mixing is not directly correlated with changes in path height. The more appropriate time scale for photochemical loss is the mean arrival time at the location(s) where each trace gas is rapidly destroyed. Once an air parcel passes through a region of rapid photochemical loss for a particular tracer the first time then all of that tracer is destroyed (converted into chlorine and bromine in this case) and it doesn't matter what happens to the air parcel from then on as far as the fractional release. Subsequent aging has no further effect on the release of chlorine or bromine. The mean arrival times at the region of rapid photochemical destruction for each trace gas will be dependent on the stratospheric transport each year and will be variable.

*This comment is to a large part in line with the comments by reviewer #1, except that here the region of rapid photochemical loss is emphasized, whereas reviewer #1 uses the maximum path height (MPH) as a proxy for this. As explained above, we do not want to state that transit time is the only factor, but we think that there are good arguments that transit time is an important factor and that **on average** the time in the rapid photochemical loss region increases with transit time. Similar as explained above we included a more detailed discussion of this in the text:*

*The fractional chemical loss can be expressed in a very generalized way as  $(1-f(t^{\wedge},p))$ , where  $f(t^{\wedge},p)$  is a fractional release function, which is specific for each trace gas and will depend on the time the air parcel has spent in the stratosphere  $t^{\wedge}$  and on the path  $p$  it has taken during the transport, especially also of the maximum path height (MPH) (Hall, 2000). While the path or the MPH are not known, it has been shown that “molecules arriving at  $X$  with long arrival times will, on average, have spent more time exposed to chemical loss and will have sampled atmospheric regions where photochemical loss is greater” (Plumb et al., 1999). We therefore follow the approach that the fractional loss can on average be described as function of the transit time only. That chemical loss and transit time are on the average related to each other is also reflected in the tight observed correlations between mean age and tracer mixing ratios (e.g. Volk et al., 1997; Engel et al., 2002). While there will be fluid elements with very different paths and different chemical loss which have the same transit time the loss can on average be sufficiently well described as a function of the transit time. We therefore treat  $f(t^{\wedge},p)$  as  $f(t^{\wedge})$  only. This is also in line with the findings of Schoeberl et al. (2000), who showed that using an “average path approximation” with a “single-path photochemistry” and thus with a unique relationship between loss and transit time, global tracer-tracer correlations can be explained. This concept that loss can be described only as a function of transit time without considering the different transit*

5 *pathways was also adopted by Schoeberl et al. (2005) in the derivation of age spectra. We use a mean age of 3 years for mid latitudes and of 5.5. years for high latitudes. Indeed, the path distribution for an air parcel with mean age of 3 years in the tropics, in mid latitudes and in polar regions is expected to show more variability than for air parcels investigated under similar conditions (e.g. latitude regions). As this analysis is restricted to one latitude band for one mean age level, we therefore approximate loss as a function of transit time only.*

2. The assumption of constant stratospheric dynamics and photochemistry from the late 20th century to the late 21st century is certainly not a good one. What is the sensitivity of the results to the predicted changes in the stratospheric circulation?

10 *If the circulation accelerates, as projected by all climate models, we expect an increase in fractional release on a given mean age surface and also an upward movement of mean age surfaces in the stratosphere. The exact extend is strongly dependent on changes in residual circulation and in mixing and this will certainly affect fractional release and thus inorganic halogen loading. We have included a comparison between our new EESC calculation and ESC from a chemistry climate model (EMAC) In the new section 4.3. Based on this we conclude that EESC is a reasonable proxy for ESC on a constant pressure level, in*  
15 *agreement with findings by Shepherd et al.*

*We have added the following in the introduction:*

20 *Note that EESC is only a valid proxy for anthropogenic ozone depletion if all other parameters, especially atmospheric transport, are unchanged. A projection of a return of EESC to some specific level therefore does not imply that ozone will return to the same levels. EESC should thus be regarded as proxy for the impact of halogenated source gases on the ozone layer due to both anthropogenic and natural emissions. The recovery of the ozone layer is affected by other parameters in addition, especially changes in transport. EESC is therefore not a proxy for ozone recovery, but a proxy for the impact due to one single parameter, the halogen loading. In section 4.3. we discuss the validity of EESC as a proxy for inorganic halogen*  
25 *loading under the influence of changing stratospheric dynamics.*

*We have further added more on this issue in the new section 4.3., especially the section on*

### ***Comparison to model calculations with varying dynamics***

30 *Under changing stratospheric dynamics (e.g. Butchart, 2014), it is expected that fractional release factors at a given mean age level will change (Douglass et al., 2008; Li et al., 2012; Ostermüller et al., 2017). Therefore, the inorganic halogen loading as a function of mean age would be expected to change even if all source gases remained constant in time. Under such conditions, EESC is not expected to follow ESC on a given mean age level. To estimate the validity of EESC as a proxy for inorganic*  
35 *halogen loading of the stratosphere, we have compared our new formulation to a free running chemistry climate model simulation. We used data from the EMAC model simulation RC2-base-04 from the ESCiMo project for this (Jöckel et al., 2016). This simulation covers the 1950–2100 time frame with simulated sea surface temperatures and sea ice contents. As described above, we again calculated fractional release factors from the model in order to have results which are internally consistent. Northern Hemisphere fractional release factors for the year 2000 are in good agreement with observation based*  
40 *fractional release factors (Newman et al., 2007; Laube et al., 2013) and therefore we used northern hemispheric data for this comparison. In addition to comparing ESC on a fixed mean age level we also compared ESC on a fixed pressure level to our new formulation of EESC. A similar comparison has been presented in Shepherd et al. (2014), who compared model ESC on a fixed pressure level to EESC on fixed mean age level (using the formulation of Newman et al. (2007)), showing good agreement. Figure 9 compares the time evolution of EESC for 3 years of mean age with model ESC at the 60 hPa level (corresponding to 3 years of mean age in the year 2000) and model ESC at 3 years of mean age. The year of 2000 and the*  
45 *corresponding level of 60 hPa was chosen, as we also evaluated fractional release factors in the year 2000 of the model run. As expected, ESC at 3 years of mean age deviates systematically from EESC, especially in the future when fractional release evaluated on a mean age surface changes significantly in the model. The agreement with ESC on a fixed pressure level is*

5 however much better. In this comparison EESC at 3 years of mean age would slightly overestimate ESC on a pressure level in the future and significantly underestimate ESC on a mean age level. The exact magnitude of changes in stratospheric dynamics is highly uncertain and also we expect ozone to follow a pressure surface rather than a mean age surface in the future. We therefore conclude that EESC is a reasonable proxy for the effect of halogen loading on stratospheric ozone, given the overall high uncertainties associated to the future evolution of stratospheric dynamics.

In the discussion section we have added the following:

10 We have shown that the long-term evolution of equivalent stratospheric chlorine (ESC, i.e. inorganic chlorine and bromine, the latter weighted in a similar way as in EESC to reflect the higher efficiency of bromine to ozone depletion) in the model deviates substantially from our calculation of EESC in a long-term model calculation with varying dynamics. However, we have also shown that the new formulation of EESC is a reasonable proxy for the evolution of inorganic halogen loading on a given pressure level. We therefore conclude that EESC is a reasonable proxy for future halogen impact on ozone.

15 As current climate models consistently predict an acceleration in the Brewer-Dobson circulation (Butchart, 2014), this will have an impact on the temporal evolution of inorganic halogen loading of the stratosphere. These expected changes in the Brewer-Dobson circulation would result in an earlier recovery of ozone at mid- and high latitudes (Eyring et al., 2010). These changes are not included in the concept of EESC. However, we have shown that EESC is a reasonable proxy for ESC when ESC is evaluated at constant pressure level.

20 3. N<sub>2</sub>O is only mentioned briefly in the conclusions but mid-latitude ozone depletion in the late 21st century will be due primarily to N<sub>2</sub>O concentrations (Ravishankara et al., 2009, Portmann et al., 2012, Butler et al., 2016). The variability and uncertainty in the N<sub>2</sub>O concentrations will be much more of a factor in the return of mid-latitude ozone to 1980 levels than the small variability in the decline of EESC.

30 The reviewer claims that ozone depletion in the 21st century will be primarily due to N<sub>2</sub>O concentrations. This is not correct. The paper by Ravishankara states that in the 21st century the emissions of N<sub>2</sub>O are more important than the emissions of any individual CFCs. The ozone depletion due to chlorine concentrations is nevertheless still the dominant anthropogenic influence on ozone. In addition, N<sub>2</sub>O cannot just be included in the same way as chlorine and bromine, as its efficiency for ozone depletion changes with changing halogen loading (Daniel et al., 2010). This would add another level of complexity, which is not the main focus of this study. In order to make this clear, we added the following in the introduction section of the paper.

35 While in principle, N<sub>2</sub>O could be included in EESC, as the NO<sub>x</sub> released in the stratosphere also leads to ozone depletion (Ravishankara et al., 2009), this is complicated, as the efficiency with which N<sub>2</sub>O leads to ozone depletion changes with halogen loading (Daniel et al., 2010; Ravishankara et al., 2009). We therefore do not include N<sub>2</sub>O in EESC.

40 4. All of the above references are relevant to this study and none of them were included. The study of Waugh et al. (2007) is also highly relevant since they explore and discuss the age vs. path sensitivity of inorganic chlorine in the stratosphere. A further point, what is special about recovery to 1980 levels? As was discussed in Newman et al. (2007) the year chosen to be the initial year causes large variability in the recovery time due to the steep slope of EESC around 1980 and the gradual slope in the late 21st century.

45 We thank the reviewer to pointing us to further studies and apologize for not including them. We have included more references to further work on links between dynamical changes and chemical changes and also on the factors influencing chemical loss. In the introduction we have added:

In models which include both chemistry and transport of the stratosphere, the amount of inorganic halogen can be directly calculated as Cl<sub>y</sub> and Br<sub>y</sub>. On average, the inorganic halogen content is higher for air parcels with higher mean age (Newman

*et al., 2007;Engel et al., 1997), but the relation between Cl<sub>y</sub> and mean age may differ from model to model depending on the representation of transport and chemistry in the model (Vaugh et al., 2007). The relation between mean age and Cl<sub>y</sub> depends on the interaction between transport and chemistry and is a function of both the time spent in the stratosphere and the transport pathways (Hall, 2000;Schoeberl et al., 2000;Schoeberl et al., 2005;Vaugh et al., 2007).*

5 *Further in the section 2 which describes the influence of transport and tropospheric trends on chemical active species, we have added a detailed discussion of the dependency, including the relevant references:*

*The fractional chemical loss can be expressed in a very generalized way as  $(1-f(t^{\wedge},p))$ , where  $f(t^{\wedge},p)$  is a fractional release function, which is specific for each trace gas and will depend on the time the air parcel has spent in the stratosphere  $t^{\wedge}$  and on the path  $p$  it has taken during the transport, especially also of the maximum path height (MPH) (Hall, 2000). While the path or the MPH are not known, it has been shown that “molecules arriving at X with long arrival times will, on average, have spent more time exposed to chemical loss and will have sampled atmospheric regions where photochemical loss is greater” (Plumb et al., 1999). We therefore follow the approach that the fractional loss can on average be described as function of the transit time only. That chemical loss and transit time are on the average related to each other is also reflected in the tight observed correlations between mean age and tracer mixing ratios (e.g.Volk et al., 1997;Engel et al., 2002). While there will be fluid elements with very different paths and different chemical loss which have the same transit time the loss can on average be sufficiently well described as a function of the transit time. We therefore treat  $f(t^{\wedge},p)$  as  $f(t^{\wedge})$  only. This is also in line with the findings of Schoeberl et al. (2000), who showed that using an “average path approximation” with a “single-path photochemistry” and thus with a unique relationship between loss and transit time, global tracer-tracer correlations can be explained. This concept that loss can be described only as a function of transit time without considering the different transit pathways was also adopted by Schoeberl et al. (2005) in the derivation of age spectra. We use a mean age of 3 years for mid latitudes and of 5.5. years for high latitudes. Indeed, the path distribution for an air parcel with mean age of 3 years in the tropics, in mid latitudes and in polar regions is expected to show more variability than for air parcels investigated under similar conditions (e.g.latitude regions). As this analysis is restricted to one latitude band for one mean age level, we therefore approximate loss as a function of transit time only.*

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### 25 **Reviewer #3**

The paper studies the combined effect of chemical loss and transport for the formulation of a simplified index describing the ozone depleting capacity due to halogens in the atmosphere. Usually, for that purpose the quantity EESC is used (for example in the WMO ozone assessments). EESC experienced several changes in its definition already in the past, when the correction for transport times was refined including its impact on release factors and the impact of transport time distribution when correcting for a non-linear trend has been included. Following Newman et al. (2007), the authors here present a further refinement of this concept. Generally, I find this paper a very nice exercise to better understand how chemical loss and transport work together, here applied to describe the halogen loading in the atmosphere. The value of the paper is to remove inconsistencies in the formulation of the traditional EESC concept, but still use the same simplified approach. The paper is clearly written, references are given as necessary, the assumptions are discussed in detail and possible consequences for the ozone recovery description are discussed.

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5 My main objection is the title of the paper which is in my opinion totally misleading. The paper is about a refinement of the EESC concept. In this concept, parameters can be derived like the time of recovery. This parameters has a descriptonal character, and can be used to compare different scenarios of Halogen loading, for example. Modifying the formulation of the concept will change the value of a parameter, but not the scenario, and therefore also not its "real" Halogen loading. In addition, as the authors state by themselves, the concept does not include deviations of the implicit assumed stationarity of the dynamics which is not even true for the past.

10 *We completely agree; the title was really misleading. We have renamed the paper in order to emphasize that the main goal is to derive a new formulation of EESC and that we then discuss some implications of this formulation. The title has been changed to "A refined method for calculating Equivalent Effective Stratospheric Chlorine".*

15 My second concern is somewhat related: as the EESC concept describes only one of the main driving processes for additional ozone depletion I would ask the authors therefore to put their results in the context of model studies where the effect of the accelerating Brewer Dobson circulation has been analysed.

We have now included more discussion on the link between EESC and changes in the Brewer Dobson circulation. In the introduction:

20 *We have further added more on this issue in the discussion (see also answers to reviewer 2 and 3):*

25 *Note that EESC is only a valid proxy for anthropogenic ozone depletion if all other parameters, especially atmospheric transport, are unchanged. A projection of a return of EESC to some specific level therefore does not imply that ozone will return to the same levels. EESC should thus be regarded as proxy for the impact of halogenated source gases on the ozone layer due to both anthropogenic and natural emissions. The recovery of the ozone layer is affected by other parameters in addition, especially changes in transport. EESC is therefore not a proxy for ozone recovery, but a proxy for the impact due to one single parameter, the halogen loading. In section 4.3. we discuss the validity of EESC as a proxy for inorganic halogen loading under the influence of changing stratospheric dynamics.*

30 *We have further added more on this issue in the new section 4.3., especially the section on*

### **Comparison to model calculations with varying dynamics**

35 Under changing stratospheric dynamics (e.g. Butchart, 2014), it is expected that fractional release factors at a given mean age level will change (Douglass et al., 2008; Li et al., 2012; Ostermüller et al., 2017). Therefore, the inorganic halogen loading as a function of mean age would be expected to change even if all source gases remained constant in time. Under such conditions, EESC is not expected to follow ESC on a given mean age level. To estimate the validity of EESC as a proxy for inorganic halogen loading of the stratosphere, we have compared our new formulation to a free running chemistry climate model simulation. We used data from the EMAC model simulation RC2-base-04 from the ESCiMo project for this (Jöckel et al., 2016). This simulation covers the 1950–2100 time frame with simulated sea surface temperatures and sea ice contents. As described above, we again calculated fractional release factors from the model in order to have results which are internally consistent. Northern Hemisphere fractional release factors for the year 2000 are in good agreement with observation based fractional release factors (Newman et al., 2007; Laube et al., 2013) and therefore we used northern hemispheric data for this



comparison. In addition to comparing ESC on a fixed mean age level we also compared ESC on a fixed pressure level to our new formulation of EESC. A similar comparison has been presented in Shepherd et al. (2014), who compared model ESC on a fixed pressure level to EESC on fixed mean age level (using the formulation of Newman et al. (2007)), showing good agreement. Figure 9 compares the time evolution of EESC for 3 years of mean age with model ESC at the 60 hPa level (corresponding to 3 years of mean age in the year 2000) and model ESC at 3 years of mean age. The year of 2000 and the corresponding level of 60 hPa was chosen, as we also evaluated fractional release factors in the year 2000 of the model run. As expected, ESC at 3 years of mean age deviates systematically from EESC, especially in the future when fractional release evaluated on a mean age surface changes significantly in the model. The agreement with ESC on a fixed pressure level is however much better. In this comparison EESC at 3 years of mean age would slightly overestimate ESC on a pressure level in the future and significantly underestimate ESC on a mean age level. The exact magnitude of changes in stratospheric dynamics is highly uncertain and also we expect ozone to follow a pressure surface rather than a mean age surface in the future. We therefore conclude that EESC is a reasonable proxy for the effect of halogen loading on stratospheric ozone, given the overall high uncertainties associated to the future evolution of stratospheric dynamics.

15 *In the discussion section we have added the following:*

*We have shown that the long-term evolution of equivalent stratospheric chlorine (ESC, i.e. inorganic chlorine and bromine, the latter weighted in a similar way as in EESC to reflect the higher efficiency of bromine to ozone depletion) in the model deviates substantially from our calculation of EESC in a long-term model calculation with varying dynamics. However, we have also shown that the new formulation of EESC is a reasonable proxy for the evolution of inorganic halogen loading on a given pressure level. We therefore conclude that EESC is a reasonable proxy for future halogen impact on ozone.*

...  
*As current climate models consistently predict an acceleration in the Brewer-Dobson circulation (Butchart, 2014), this will have an impact on the temporal evolution of inorganic halogen loading of the stratosphere. These expected changes in the Brewer-Dobson circulation would result in an earlier recovery of ozone at mid- and high latitudes (Eyring et al., 2010). These changes are not included in the concept of EESC. However, we have shown that EESC is a reasonable proxy for ESC when ESC is evaluated at constant pressure level.*

30 The derivation of the concept is somewhat lengthy in my opinion and can be combined, for example for eqs. (11)-(14). Stationarity means that at the end that the combined history of an air parcel (including its mean photochemical dose) is only a function of the position in the atmosphere. So equation (19) is not a surprise. Much more subtle is the transition from equ. (19) to (20). This is valid only under the specific condition that the distribution  $G\#$  is determined by its first moment only which may be not generally true. Here I would ask the authors to discuss the assumptions in more detail.

35 *As none of the other reviewers has commented on this derivation being too long, we would rather keep it in its present form. The transition from (19) to (20) does indeed imply that all locations where mean release time has the same value, the release time distribution is the same. This is very closely linked to the discussion of path dependency (see answers to reviewer #1 and #2 and general introduction to our answers). In order to make this underlying assumption we have included the following statement in the explanation of the transition from (19) to (20):*

*This implies that at all locations  $r$  with the same mean release time, the release time distribution is the same. This assumption may not be valid everywhere, but as a mean age of 3 years is used for mid latitudes and of 5.5 years for high latitudes, we use this assumption only for air parcels under similar meteorological conditions (latitude bands).*

- 5 Finally, to be more than just an interesting exercise, the paper would strongly improve if the authors could show that using their new formulation would yield a more concise ozone trend analyses, at least in one example

*It is not the intention of this paper to repeat studies using EESC as a proxy for halogen-induced ozone loss. In any case, as the timing of e.g. the maximum is rather similar in both formulations, we do not believe that the results would be very different.*

- 10 In order to emphasize this we have included the following in the conclusions:

*As the suggested reformulation of EESC does not affect the principal behavior of the temporal evolution of EESC, we do not expect this reformulation to lead to substantial changes, which could impact the changes of studies using EESC except for those which have used EESC to project EESC recovery.*

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### **Minor points**

p219: transport within: *changed to into and within*

- 20 p4117: you mean averaged over the seasons == annual mean: *seasonal mean changed to annual mean*

p4118: eliminate "it is expected": *deleted*

- 25 p4120: the typical path from the tropical tropopause through the stratosphere back to the troposphere at higher latitudes will not yield this shorter lifetime at the end of the path. You mean strictly in the stratosphere.

*Yes, and actually this should also be restricted to the tropical latitudes. We added: "and topical latitudes in the stratosphere"*

- 30 p4121:  $f$  will be a function of  $r$ , too. This does not harm the derivation.

*This is the linked to the main point raised by reviewer 1 and 2. We added a discussion on the additional dependence on the location, or more specifically the transport pathway. Concerning the dependence on the location, we have added the following:*

- 35 *The fractional chemical loss can be expressed in a very generalized way as  $(1-f(t^{\wedge},p))$ , where  $f(t^{\wedge},p)$  is a fractional release function, which is specific for each trace gas and will depend on the time the air parcel has spent in the stratosphere  $t^{\wedge}$  and on the path  $p$  it has taken during the transport, especially also of the maximum path height (MPH) (Hall, 2000). While the path or the MPH are not known, it has been shown that "molecules arriving at  $X$  with long arrival times will, on average, have spent more time exposed to chemical loss and will have sampled atmospheric regions where photochemical loss is greater" (Plumb et al., 1999). We therefore follow the approach that the fractional loss can on average be described as function of the transit time only. That chemical loss and transit time are on the average related to each other is also reflected in the tight observed correlations between mean age and tracer mixing ratios (e.g. Volk et al., 1997; Engel et al., 2002). While there will be fluid elements with very different paths and different chemical loss which have the same transit time. on the average the loss can be sufficiently well described by the transit time. We therefore treat  $f(t^{\wedge},p)$  as  $f(t^{\wedge})$  only. This is also in line with the findings of Schoeberl et al. (2000), who showed that using an "average path approximation" with a "single-path photochemistry" and thus with a unique relationship between loss and transit time, global tracer-tracer correlations can be explained. This concept that loss can be described only as a function of transit time without considering the different transit pathways was also adopted by Schoeberl et al. (2005) in the derivation of age spectra. We use a mean age of 3 years for mid latitudes and of 5.5. years for high latitudes. Indeed, the path distribution for an air parcel with mean age of 3 years in the tropics, in mid latitudes and in polar regions is expected to show more variability than for air parcels investigated under*

*similar conditions (e.g. latitude regions). As this analysis is restricted to one latitude band for one mean age level, we therefore approximate loss as a function of transit time only.*

p5131: what are the three? I see the trend and the chemical loss only.

5

*The three function are the age spectrum, the loss function and the temporal trend. We have changed the order slightly to have the three functions immediately after each other:*

*... must be considered, which are all functions of the transit time. We will denote transit time, i.e. the time a fluid element has spent in the stratosphere as  $t'$ , while the time itself will be denoted as  $t$ . First, the transit time distribution, i.e. how long it has taken for the individual fluid elements of this air parcel to travel from their entry point to the stratosphere to the location  $r$  in the stratosphere. Second, the temporal trend of the mixing ratios at the entry point has to be considered and, third, chemical loss during this transport.*

10

p612: the exponential chemical loss term is only used here and can be left out.

15 *Yes, we have left out the exponential term and used the fractional lost directly.*

*... and the chemical loss term, which can be described by the factor  $(1 - f(t'))$ , where  $f(t')$  describes the fraction which has been lost.*

$$\chi_{strat}(r, t) = \int_0^{\infty} \chi_0(t - t') \cdot (1 - f(t')) \cdot G(r, t') dt' \quad (6)$$

*As  $(1 - f(t'))$  is the remaining fraction of the organic .....*

20

p1119 remove "classical": *removed*

Typos:

parameterization/parameterization should be typed in one version only

25

*all changed to parameterization*

p319: the first moment has a lower value: *changed*

### 30 **Reviewer #4**

This manuscript has built nicely on Ostermoller et al. (2017). The concept developed there is used to derive a relationship between previously calculated fractional release (FRF) values that assumed an age spectrum representative of an inert tracer to FRF values that are independent of tropospheric source gas trends. More importantly, this work quantifies the importance of using an age spectrum that accounts for chemical loss when calculating equivalent effective stratospheric chlorine (EESC). This improved approach effectively leads to older air in the EESC calculation, particularly for the midlatitude stratosphere. This, in turn, implies lower EESC values in 1980; this 1980 level has been important because it has typically been taken as a value of significance in the return of stratospheric chlorine/bromine to natural levels. The proposed EESC revision (i.e., older air) also leads to higher EESC values for any given time when source gases are declining. These changes combine to lead to a substantial delay in the time when mid-latitude EESC is projected to return to 1980 levels. As expected, the effects are smaller for polar EESC, since the difference in the average age for the dissociated ODSs and an inert tracer are much reduced.

40

I have a few general comments here, and some more specific ones below. Assuming these comments can be dealt with sufficiently, I find this manuscript to be valuable and I believe that it should offer an important improvement on work that came before it. It would be useful to describe whether EESC from the new formalism is distinct enough from EESC using the old one so that past work that used EESC should have identified a shortcoming in the previous approach. Looking at Figure 3, I would be particularly interested in previous work that compared measurements or model calculations over a time range that spanned both before and after the EESC peak in the late 1990's, since the differences should be most apparent over such a period. If the two approaches are not distinct enough to be apparent in previous work, this would be worth stating here, so the reader knows the main impact is on the "recovery" date, and that it doesn't affect the validity of previous results.

10 *As the timing of e.g. the maximum is quite similar in both formulations, we do not believe that the results would be very different. In order to emphasize this we have included the following in the conclusions:*

15 *As the suggested reformulation of EESC does not affect the principal behavior of the temporal evolution of EESC, we do not expect this reformulation to lead to substantial changes, which could impact the changes of studies using EESC except for those which have used EESC to project EESC recovery.*

The only other comment I particularly want to highlight here relates to the sensitivity study of the width of the transport distribution function. Please see my comment below for page 14, lines 2-3. I would find this most useful if you explored the impact of a change in width of the age distribution relevant to an inert tracer, with that impact propagating to the halocarbons depending on their chemical loss; however, unless I am mistaken, it doesn't seem like this is what is done.

*See our reply to this specifically below (answer to comment on p 14, l.2-3.)*

Specific comments:

25 Page 1, Line 1 At some point, relatively early in the manuscript, you should make clear what you are not implying by this title, otherwise it could be considered misleading. As currently written, it could be taken to suggest that there has been more ODS emission than expected or that dynamics may change in an unexpected way to alter halogen loading in the future. An alternative that may be preferable would be to change to a title more focused on the delay in EESC recovery.

30 *As noted by all reviewers (and we completely agree), the title was misleading. The new title "A refined method for calculating Equivalent Effective Stratospheric Chlorine" does not put the focus on an estimated recovery date anymore.*

1, 16 1980 is not the year of stratospheric ozone depletion onset, but it is often used as a benchmark to measure significant progress towards recovery

35 *Rephrased to "used a benchmark to which EESC must return in order to reach significant progress towards halogen and ozone recovery."*

3,16 I suggest clarifying what 'this purpose' refers to at the end of this sentence

40 *The sentence has been changed to clarify this: "The age spectrum for in inert tracer is not well suited to describe the propagation of a tracer with chemical loss into the stratosphere."*

5,1-3 It is not clear to me that this sensitivity study addresses the entire phase space of possibilities in your assumed relationship between age and loss. Additional justification is needed to show that the simple relationship you are basing your calculations on are sufficiently appropriate.

50 *The Figure showing the different transit time distributions is purely for illustrative purposes and makes no claim that these are the real distributions. As explained in the manuscript (section 4), we have varied several parameters in order to test the influence on EESC calculated using our new formulation and found that the impact on EESC and on projected recovery dates*

is not very large. The best way to show that the relationship used here is sufficiently appropriate would be to compare EESC with **observations** of inorganic chlorine and bromine. (Plumb et al., 1999) showed that it gives very much improved representation of organic species, which is a good indication that the inorganic fraction should also be better described. Such observations of inorganic chlorine and bromine are not available, which is why we have chosen to test our new formulation in a full model calculation. A new section on this comparison has been added (section 4.3., especially the first part on the comparison with a fixed dynamics model calculation), which shows that the new formulation yields much improved agreement with model calculations, although certain discrepancies remain. We have also added in the conclusions to the paper that the relationship between loss/release and transport should be explored in more realistic models.

10 *Such calculations are only available based on a rather old 2D model (Plumb et al., 1999) and should be repeated with state-of-the-art models.*

6, 2 It is not clear to me that having the loss described as an exponential term with the lifetime depending on transit time is helpful in the formulation. It is really of an arbitrary mathematical form since the lifetime (denominator) varies with the location endpoint. It would seem more straightforward to skip straight to the factor  $(1-f(t'))$ , but I leave this decision to the authors.

*Yes, the exponential function has been deleted and we now go straight to the factor  $(1-f(t'))$*

20 10, 19 Somewhere you should discuss the impact of using the Plumb age estimates from an old 2-D model given the advancements in our ability to calculate circulation metrics over the last 20 years and the general superiority of 3-D models at making these calculations today

25 *The reviewer is correct in pointing this out. We had included in the conclusions, that the parameterizations should be revisited with state of the art models. In order to make this clearer, we have added a statement on this in the new subsection **Sensitivity to the mean release time derived from mean age and stratospheric lifetime:***

30 *Despite this rather low sensitivity, it should be noted that the parameterization is derived from a 2D model. The relationship between mean age, age spectrum and chemical loss should be explored in state-of-the-art 3 D models, which have a better representation of stratospheric transport processes.*

*In the place highlighted by the reviewer, we have added the following:*

35 *Plumb et al. (1999) used a 2D model for their study. Despite this, the stratospheric lifetimes derived from the model are in overall good agreement to more recent model studies (Chipperfield et al., 2013). The sensitivity of the parameterization between mean release time and mean age to the stratospheric lifetimes is further discussed in section 4.*

40 14, 2-3 I am having trouble understanding exactly what is being done here. Are the factors changed for both G and G<sub>N</sub>? It would not be possible to have gamma be 0 for G<sub>N</sub> and be 0.7 for G, would it? But if both gamma factors are 0, it would seem that this approach would collapse to the old result since the mean release age would be the same as the mean age of an inert species. And if that is the case, I would have expected a larger impact on return times (i.e., they should be close to the VD (2014) values). Perhaps it would help if you had a figure (like Figure 1) showing what the G curves look like as gamma goes to 0 and for it equal to 2. It looks like you may be using the values from Table 1; however, this doesn't seem appropriate if you want to examine the impact of a changing shape in the overall transport distribution function. In fact, I'm unclear physically what is going on here, so clarification would be very helpful.

45 *Thank you for this suggestion, which we have considered. As Figure 1 is however only for illustrative purposes and does not claim to be represent the real loss time and arrival time distributions, we think it would give too much weights to this Figure if we included it for other parameterizations. Therefore, we would rather not include additional similar Figures.*

50 *Concerning the comment on the range of lamda factors (we think this is what the reviewer is commenting on, not gamma values, as we varied lambda between 0 and 2) included in the sensitivity study, the reviewer is correct, that in case of a gamma*

value of 0 for the age spectrum, the mean release time would be the same as the mean age. We did not apply this sensitivity study to the derivation of mean arrival time from mean age. This sensitivity is investigated by varying the assumed stratospheric lifetimes. The sensitivity test described here is only applied to  $G_N^\#$ . In the new method for the calculation of EESC there is actually no  $G$  anymore, just the  $G_N^\#$  distributions for the various tracers. So it is only these distributions which are varied.

5 Indeed, a gamma factor of 0 for  $G_N^\#$  does not make a lot of sense, since this would mean that there is no mixing at all and this would make the whole concept of an age spectrum obsolete. But it could be regarded as a limiting case: the age spectrum cannot possible be any more narrow than this, as a gamma factor of 0 is equivalent to a pure propagation without mixing. On the other hand, a gamma value of 2 is equivalent to a very wide spectrum. Therefore, the tested values do indeed represent extreme cases and the purpose was to show that the sensitivity to the exact shape of  $G_N^\#$  is rather low, but that the main sensitivity is to the first moment of the distribution, which is the mean release time. We have broken down this subsection into two subsections, one dealing with the sensitivity to the shape of the release time distribution and one dealing with the sensitivity due to the derivation of the mean release time. These section are now called “Sensitivity to the shape of the new release time distribution  $G_N^\#$ ” and “Sensitivity to the mean release time derived from mean age and stratospheric lifetime”. To specify that we only varied the shape of  $G_N^\#$ , we have added this in the title of the subsection “Sensitivity to the shape of the new release time distribution  $G_N^\#$ ” and also in the text::

... values of 0 and 2 years in the calculation of  $G_N^\#$ , while retaining the first moment, i.e.  $\Gamma^\#$ . The ..

20 14, 25-29 Please describe how the destruction vs. age relationship is determined for these perturbations

The perturbations are simply introduced by changing the stratospheric lifetime in the formula given by Plumb et al., for the calculation of mean arrival time from mean age and stratospheric lifetime. We have added a further description to make this clear:

25

We tested the sensitivity of our calculation to that by systematically increasing all lifetimes by 20% or decreasing them by 20% (see Table 5) in the parameterization given by (Plumb et al., 1999). This results in different mean arrival time  $\Gamma^*$  and mean release time  $\Gamma^\#$  .:

30 15, 15 Perhaps broaden this statement, if you think it is accurate to something like “This approach more accurately represents the amount of Cly and Bry in the stratosphere from tropospheric source gas concentrations, and should be adopted to estimate. . .” This would seem to be more consistent with the title, but do please refer to my earlier concerns of such a broadening.

35 As noted before, we have changed the title of the paper. Nevertheless we agree with the reviewer and have added the suggested statement.:

... calculated by the method of Ostermüller et al. (2017). This approach more accurately represents the amount of  $Cl_y$  and  $Br_y$  in the stratosphere from tropospheric source gas concentrations and fractional release factors. We suggest it should be adopted to calculate EESC and to estimate ...

40

Minor comments:

1, 10 Do you mean ‘adopted’ here? While it is adapted through your work, that doesn’t seem to be the intent here

Changed to adopted

45

1, 18 Change ‘assumed’ to ‘estimated’

Changed to estimated

50 1, 27 Change to ‘winter and springtime’: changed

2,1 Replace ‘effectiveness’ with ‘extent’ or something similar; otherwise it could sound like the destruction per Cl molecule is what you are referring to here

5 *Changed to extent*

2,2 Reference EESC: *done, referenced Daniel and Velders 2011 and Newman et al., 2007.*

9, 1 Change ‘was’ to ‘way’: *changed*

10

9, 3 1-f(t) doesn’t seem to be appropriately named as the ‘loss term’; I understand why you called it the ‘chemical loss term’ back in eq. (6), but now in isolation it seems confusing that it is a ‘loss term’ that is really equal to one minus the fractional loss

15 *We are not sure if we understand this comment correctly. We refer to  $f(t)$  as the **release** (not loss) term and to  $1-f(t)$  as the loss term. We believe that this is correct.*

12, 5 Change ‘compares’ to ‘compare’: *changed*

20 12, 27 Change ‘on the new’ to ‘of the new’ at the end of the line: *changed*

13, 12 Add space in ‘STRATcampaign’: *done*

25 13, 23-24 At first, this sentence seemed to suggest that you were doing another calculation from the Ostermoller results, but in fact, I believe you are summarizing the 2060 vs 2058 discussed at the top of this page. This could be clarified.

*Upon rereading, we agree. We deleted this sentence as it only repeats what is said before and adds more confusion than clarification.*

30 14, 18 Change to ‘independent of’;: *changed*

15, 5 I don’t understand the use of the word ‘respective’ here: *changed to organic or inorganic*

35 15, 8 This seems to not be an appropriate use of ‘Therefore’. From what is stated here, the following sentence doesn’t seem to logically follow the previous statement(s).

*We have rephrased as follows for more clarity:  
Fractional release factors which are independent of the tropospheric trends (Ostermüller et al., 2017) must be used to correctly*

40 15, 16/17 I suggest changing to present tense: *done*

15, 20 Change to ‘. . . perturbed values of stratospheric chlorine and bromine. . .’ changed to “... the mid-latitude lower stratosphere to unperturbed values of chlorine and bromine” ...

45 15, 21-23 You should probably also point out that CO<sub>2</sub> is expected to accelerate column ozone recovery across much of the globe (see, e.g., Butler et al., 2016 and many others)

The reviewer is referring to the expected acceleration of the Brewer-Dobson circulation due to climate change. We have given this issue much more weight in several parts of the manuscript. We have further added a statement on ozone recovery here, referencing a paper on ozone recovery from many models (Eyring et al., 2010):

- 5 *These expected changes in the Brewer-Dobson circulation would result in an earlier recovery of ozone at mid- and high latitudes (Eyring et al., 2010). In addition to this, increases in the concentrations of N<sub>2</sub>O and short-lived chlorine containing halocarbons may further influence the recovery of the ozone layer, possibly leading to a later recovery (Hossaini et al., 2015b; Hossaini et al., 2015a; Chipperfield, 2009).*
- 10 15, 33 It could be useful to say what G# is here, so people who didn't read the main text will know what it is.  
"of the loss weighted transit distribution" has been added as explanation
- 16, 10 Make 'distributions' singular. *done*
- 15 16, 17 Change 'calculation' to plural. *done*
- Table 1 For CFC-113 and CH<sub>3</sub>Br add another significant figure '0' to the end of the time-independent FRF. Same comment for various species in Table 2 in last 2 columns
- 20 *done*
- 21, 2 Perhaps add 'the' between 'In' and 'case': *done*
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# ~~Delayed Recovery of mid-latitude lower stratospheric Halogen Loading~~A refined method for calculating Equivalent Effective Stratospheric Chlorine.

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- Abstract.** Chlorine and bromine atoms ~~can~~ lead to catalytic ~~destruction-depletion~~ of ozone in the stratosphere. Therefore the use and production of ozone depleting~~-~~substances (ODS) containing chlorine and bromine is regulated by the Montreal Protocol to protect the ozone layer. Equivalent Effective Stratospheric Chlorine (EESC) has been ~~ad~~apted as an appropriate metric to describe the combined effects of chlorine and bromine released from halocarbons on stratospheric ozone. Here we revisit the concept of calculating EESC. We derive a ~~new-refined~~ formulation of EESC based on an advanced concept of ODS propagation into the stratosphere and reactive halogen release. A new transit time distribution is introduced in which the age spectrum for an inert tracer is weighted with the release function for inorganic halogen from the source gases. This distribution is termed the “release time distribution”. ~~We show that a much better agreement to inorganic halogen loading from the chemistry transport model TOMCAT is achieved than using the current formulation.~~ The ~~refined-improved~~ formulation shows ~~that~~ EESC levels in the year 1980 for the mid latitude lower stratosphere, ~~which are-were~~ significantly lower than previously calculated. 1980 marks the year commonly ~~defined-used~~ as a benchmark to which EESC must return in order to reach ~~significant progress towards halogen and ozone recovery, as the onset of anthropogenic ozone depletion in the stratosphere.~~ Assuming that ~~- under otherwise unchanged conditions -~~ the EESC value must return to the same level in order for ozone to fully recover, we show that it will take more than 10 years longer than ~~currently-assumed-estimated~~ in this region of the stratosphere ~~with the current method for calculation of EESC.~~ ~~Based on the improved formulation, EESC level at mid latitudes will reach this landmark only in 2060.~~ We also present a range of sensitivity studies to investigate the effect of changes and uncertainties in the fractional release factors and in the assumptions on the shape of the release time distributions. ~~We conclude that, under the assumptions that all other atmospheric parameters like stratospheric dynamics and chemistry are unchanged, the recovery of mid-latitude stratospheric ozone would be expected to be delayed by about a 10 years, in a similar way as EESC.~~ We further discuss the value of EESC as a proxy for future evolution of inorganic halogen loading under changing
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atmospheric dynamics using simulations from the EMAC model. We show that while the expected changes in stratospheric transport lead to significant differences between EESC and modelled inorganic halogen loading at constant mean age, EESC is a reasonable proxy for modelled inorganic halogen on a constant pressure level.

## 1 Introduction

5 It is well established that chlorine and bromine atoms in the stratosphere enhance ozone ~~depletion-loss~~ via catalytic reaction chains (Stolarski. and Cicerone, 1974;Solomon, 1999;Molina and Rowland, 1974;Wofsy et al., 1975). Ozone ~~loss-depletion~~ has been observed at mid latitudes (S. Pawson and W. Steinbrecht et al., 2014) and in particular at high latitudes during winter and spring time (Farman et al., 1985;M. Dameris and S. Godin-Beekmann et al., 2014). The chlorine and bromine atoms responsible for the ozone depletion are not injected directly into the stratosphere, but are released from organic halocarbons, so called ozone depleting substances (ODS), which are emitted in the troposphere. Ozone is thus not depleted by reactions with the chemicals emitted, ~~but~~ by reaction with the inorganic halogen released from these chemicals. The ~~effectiveness~~ extent of the catalytic ozone ~~destruction-depletion~~ depends on the amount of inorganic halogen in the stratosphere. In models which include both chemistry and transport of the stratosphere, the amount of inorganic halogen can be directly calculated as  $Cl_v$  and  $Br_v$ . On average, the inorganic halogen content is larger for air parcels with higher mean age (Newman et al., 10 2007;Engel et al., 1997), but the relation between  $Cl_v$  and mean age may differ from model to model depending on the representation of transport and chemistry in the model (Waugh et al., 2007). The relation between mean age and  $Cl_v$  depends on the interaction between transport and chemistry and is a function of both, the time spent in the stratosphere, and the transport pathways (Hall, 2000;Schoeberl et al., 2000;Schoeberl et al., 2005;Waugh et al., 2007). Equivalent Effective Stratospheric Chlorine (EESC) is a metric describing the combined effect of all chlorinated and brominated ODSs expressed as the equivalent amount of inorganic chlorine in the stratosphere based on tropospheric abundances of tropospheric source gases (Daniel and Velders, 2011;Newman et al., 2007). While in principle,  $N_2O$  could be included in EESC, as the  $NO_x$  released in the stratosphere also leads to ozone depletion (Ravishankara et al., 2009), this is complicated, as the efficiency with which  $N_2O$  leads to ozone depletion changes with halogen loading (Daniel et al., 2010;Ravishankara et al., 2009). We therefore do not include  $N_2O$  in EESC. EESC depends on the transport from the troposphere into the stratosphere, the temporal trend of the mixing ratios of the source gases in the troposphere and the release of inorganic halogen from these source gases. EESC has been used widely as a proxy to describe the combined effects of inorganic bromine and chlorine on stratospheric ozone, e.g. in the analysis of time series of ozone or when discussing the effects of volcanoes or geoengineering (Tilmes et al., 2009;Shepherd et al., 2014;Weatherhead and Andersen, 2006;Chipperfield et al., 2017). Note that EESC is only a valid proxy for anthropogenic ozone depletion, if all other parameters, especially atmospheric transport, are unchanged. A projection of a return of EESC to some specific level therefore does not imply that ozone will return to the same levels. EESC should thus be regarded as proxy for the impact of halogenated source gases on the ozone layer due to both anthropogenic and natural emissions. The recovery of the ozone layer is affected by other parameters in addition, especially changes in transport and 25 30

temperature. EESC is therefore not a proxy for ozone recovery, but a proxy for the impact due to one single parameter, the halogen loading. In section 4.3. we discuss the validity of EESC as a proxy for inorganic halogen loading under the influence of changing stratospheric dynamics.

The transport into and within the stratosphere is described by the mean age of air,  $\Gamma$  (Hall and Plumb, 1994; Waugh and Hall, 2002; Kida, 1983). A stratospheric air parcel does not have a single transit time  $t'$  since its entry into the stratosphere, but is rather composed of a large number of irreversibly mixed fragments or fluid elements with varying transit times  $t'$ , describing the variable times they already spent in the stratosphere. The distribution of transit times is called the age spectrum, the arithmetic mean (first moment) being the mean age  $\Gamma$ . The age spectrum is generally described by a Green's function  $G$  for one-dimensional advective diffusive transport and a parameterization of the width of the distribution as a function of the mean age (Hall and Plumb, 1994). Together with the temporal trend of the trace gas in the troposphere, the age spectrum determines the mixing ratio of an inert trace gas in the stratosphere (1) at a certain time  $t$  and place  $r$ ,  $\chi_{inert, strat}(r, t)$ , as the fluid elements will each contain the mixing ratio present in the troposphere at the time they entered the stratosphere,  $\chi_0(t - t')$ . Based on this concept, it is also possible to derive mean age of air (Hall and Plumb, 1994; Volk et al., 1997; Engel et al., 2002; Engel et al., 2009) based on observations of chemically inert tracers in the stratosphere, so called age tracer.

$$\chi_{inert, strat}(r, t) = \int_0^{\infty} \chi_0(t - t') \cdot G(r, t') dt' \quad (1)$$

For chemically active species, in addition to the transport, the chemical loss leading to the release of inorganic halogen needs to be considered. This release of inorganic chlorine and bromine from the halocarbon source gases is characterized by the fractional release factor  $f$  (FRF). The FRF describes which fraction of the source gas molecules originally present in an air parcel has already been released, i.e. transferred to the inorganic fraction.  $1-f$  will thus describe the fraction that is still in the form of the organic source gas. Fractional release factors for many relevant trace gases have been determined as a function of mean age (Newman et al., 2007; Laube et al., 2013). Typically, a mean age value of 3 years is adapted for the lower stratosphere of the middle latitudes and a mean age value of 5.5 years is used for polar winter conditions in the lower stratosphere (Newman et al., 2007). The calculation of  $f$  relies on the difference of the observed mixing ratio of the source gases in the stratosphere to the amount of source gas originally present in this air-parcel (2). For this, a reference mixing  $\chi_{ref}(\Gamma)$  ratio must be determined, based on temporal trends in the troposphere and transport into the stratosphere.

$$f(\Gamma) = \frac{\chi_{ref}(\Gamma) - \chi_{strat}(\Gamma)}{\chi_{ref}(\Gamma)} \quad (2)$$

This reference mixing ratio has typically been calculated using (1), i.e. assuming that the chemically active gas propagates in the same way as a chemically inert gas. Plumb et al. (1999) showed that the age spectrum  $G$ , which is representative for an inert gas is not well suited to describe the way that a chemical active gas is propagated into the stratosphere. The reason for this is that the remaining organic fraction of a chemically active species (CAS) is determined largely by the fluid elements

with shorter transit times, where chemical loss is less pronounced. The fluid elements with longer transit times on the other hand do not contribute as much to the remaining organic fraction, as more chemical loss has occurred. The combination of chemical loss and transport is described by a modified age spectrum, called the arrival time distribution. This arrival time distribution is weighted stronger at shorter transit times and has a different first moment than the age spectrum for an inert tracer. This first moment of the arrival time distribution ~~is shorter~~ has a lower value than the mean age and is termed the mean arrival time. Information on the mean arrival time was derived from 2-D model calculations by Plumb et al. (1999), who used the mean arrival time to detrend stratospheric correlations. They found that the detrended correlations from different years showed good agreement, if the mean arrival time was used in calculating the reference values, while this was not the case when using mean age. Ostermüller et al. (2017) could show that the arrival time distribution also allows to derive fractional release factors, which are not influenced by the tropospheric trend. These studies show that, due to the interaction of chemistry and transport, changes in the tropospheric mixing ratios of source gases with chemical loss are reflected faster in their stratospheric mixing ratios than are changes in gases without chemical loss. The age spectrum for an inert tracer is not well suited for this purpose to describe the propagation of a tracer with chemical loss into the stratosphere.

EESC is influenced by the temporal trends of the source gases, their fractional release factors and the transport into the stratosphere. While all these factors may vary with time, e.g. due to changes in stratospheric circulation (Douglass et al., 2008; Li et al., 2012b), especially fractional release factors should not depend on the tropospheric trends of the trace gases (Ostermüller et al., 2017). As in the case of FRF, EESC is usually calculated as a function of mean age and again a mean age value of 3 years is adapted for the lower stratosphere of the middle latitudes and a mean age value of 5.5 years is used for polar winter conditions in the lower stratosphere (Newman et al., 2007). The formulation which is currently used to calculate EESC is based on the concept of fractional release and mean age, using the age spectrum  $G$  for an inert tracer. In this formulation EESC is calculated by multiplying the fractional release factor with the integral over the tropospheric time series  $\chi_0$  of the tracer and the age spectrum for an inert tracer (Newman et al., 2007; Velders and Daniel, 2014)

$$EESC_{current}(\Gamma, t) = \sum_{Cl} \left( n_i f_i(\Gamma) \int_0^{\infty} \chi_{0,i}(t-t') G(\Gamma, t') dt' \right) + \alpha \sum_{Br} \left( n_i f_i(\Gamma) \int_0^{\infty} \chi_{0,i}(t-t') G(\Gamma, t') dt' \right) \quad (3)$$

with  $n_i$  being the number of chlorine or bromine atoms in species  $i$  and  $f_i$  being the fractional release factor.  $\alpha$  is a factor representing the higher effectivity of bromine to ozone ~~destruction~~ depletion, typically taken as 60 for both high latitudes and mid latitudes (Newman et al., 2007). The age spectrum  $G$  used here is that for an inert tracer. As shown by Plumb et al. (1999) and Ostermüller et al. (2017), the arrival time distribution is better suited to describe the propagation of the organic fraction of a source gas into the stratosphere. Consequently, it is also expected that the age spectrum for an inert tracer may not be the best way to describe the propagation and release of the inorganic fraction and thus EESC.

In this paper we discuss the interaction of chemistry and transport in the propagation of chemically active tracers with tropospheric trends into the stratosphere and suggest an improved method for the calculation of EESC. The paper is organized as follows. In section 2 we present some general thoughts on the propagation of tropospheric trends taking into account chemical loss. In section 3 we derive a new mathematical formulation for EESC, based on the ideas developed in section 2.

5 This new mathematical formulation is applied in section 4 to the scenario of source gas mixing ratios given ~~by~~ Velders and Daniel (2014) and the results are compared to their results for the estimated recovery of EESC to 1980 values. We also present sensitivity studies of the new EESC formulation to different parameters and compare the different formulations of EESC to simulations of inorganic halogen loading from two different comprehensive three-dimensional atmospheric chemistry models. Finally we draw some conclusion and present an outlook in section 5.

10 (~~Schoeberl et al., 2005~~)

## **2. On the ~~influence of interaction between transport~~transport and tropospheric trends on chemical active species, chemistry and tropospheric trends.**

In addition to transport and temporal trends in the troposphere, the stratospheric mixing ratio of a species with chemical loss in the stratosphere depends on the loss processes and on the interplay between transport, chemical loss and the temporal trend  
15 (Volk et al., 1997;Plumb et al., 1999). The age spectrum  $G$ , which is used to describe the propagation of chemically inert trace gases into the stratosphere (Schauffler et al., 2003;Newman et al., 2007;Engel et al., 2002) and to calculate mean age does not take into account chemical loss. Chemical loss is not uniform throughout the stratosphere, as it depends in most cases on the actinic flux at short wavelength. In the ~~seasonal-annual~~ mean, the chemical lifetime will in general decrease with altitude and increase with increasing latitude(~~Douglass et al., 2008~~), leading to a clear relation between the maximum altitude of a fluid  
20 element and the fractional release (Douglass et al., 2008;Hall, 2000).~~;~~ The chemical loss is thus very inhomogeneous, but on average, ~~it is expected that~~ the longer a fluid element remains in the stratosphere, the larger the integrated chemical loss will be (Plumb et al., 1999). Also, as the loss for most species with photochemical sinks mainly occurs at higher altitudes and tropical latitudes in the stratosphere, it is expected that, on average, longer transit times will be associated with shorter lifetimes. A transit time distribution in which the transit times are weighted with the transit time dependent chemical loss has been termed  
25 “arrival time distribution” (Plumb et al., 1999),  $G^*(4)$ . The fractional chemical loss can be expressed in a very generalized way as  $(1 - f(t', p))$ , where  $f(t', p)$  is a fractional release function, which is specific for each trace gas and will depend on the time the air parcel has spent in the stratosphere  $t'$  and on the path  $p$  it has taken during the transport, especially also of the maximum path height (MPH) (Hall, 2000).~~;~~ While the path or the MPH are not known, it has been shown that “molecules arriving at X with long arrival times will, on average, have spent more time exposed to chemical loss and will have sampled  
30 atmospheric regions where photochemical loss is greater” (Plumb et al., 1999). We therefore follow the approach that the fractional loss can on average be described as function of the transit time only. That chemical loss and transit time are on the average related to each other is also reflected in the tight observed correlations between mean age and tracer mixing ratios

(e.g. Volk et al., 1997; Engel et al., 2002). While there will be fluid elements with very different paths and different chemical loss which have the same transit time the loss can on average be sufficiently well described as a function of the transit time. We therefore treat  $f(t', p)$  as  $f(t')$  only. This is also in line with the findings of Schoeberl et al. (2000), who showed that using an “average path approximation” with a “single-path photochemistry” and thus with a unique relationship between loss and transit time, global tracer-tracer correlations can be explained. This concept that loss can be described only as a function of transit time without considering the different transit pathways was also adopted by Schoeberl et al. (2005) in the derivation of age spectra. We use a mean age of 3 years for mid latitudes and of 5.5 years for high latitudes. Indeed, the path distribution for an air parcel with mean age of 3 years in the tropics, in mid latitudes and in polar regions is expected to show more variability than for air parcels investigated under similar conditions (e.g. latitude regions). As this analysis is restricted to one latitude band for one mean age level, we therefore approximate loss as a function of transit time only. Schoeberl et al. (2000) Schoeberl et al. (2005) of course a function of transit time. The first moment of the arrival time distribution ~~arrival time distribution~~ is called the mean arrival time  $\Gamma^*$ . This distribution describes the probability distribution for organic source gas molecules to arrive at some place  $r$  in the stratosphere, as a function of transit time  $t'$ :

$$G^*(r, t') \equiv (1 - f(t')) \cdot G(r, t'). \quad (4)$$

We can now define a second transit time distribution,  $G^\#(r, t')$ , which describes the probability for an inorganic halogen atom released from this source gas to arrive at this place  $r$  in the stratosphere, again as a function of transit time  $t'$ :

$$G^\#(r, t') \equiv f(t') \cdot G(r, t'). \quad (5)$$

For the calculation of  $G^*$  and  $G^\#$  the integrated loss as a function of transit time needs to be known. Purely for illustrative purposes, we have constructed such a loss function using a sigmoid function, which changes from 0 (no loss) for short transit times to 1 (complete loss) for longer transit times. The function has been constructed in a way to match fractional release factors for CFC-11 for 3 and 5.5 years. CFC-11 is one of the most important chlorine source gases for the stratosphere. These three different transit time distributions,  $G$ ,  $G^*$  and  $G^\#$  calculated using typical age spectra for 3 and 5.5 years of mean age are shown in Figure 1 and Figure 2. In both cases, the mean transit time for the inorganic fraction is longer than that for an inert tracer, while the lag of the remaining organic fraction is shorter than that of an inert tracer. The effect is much more pronounced for the 3 years mean age calculation, where the fractional release is about 0.5. i.e. the organic and the inorganic fraction are about equal. In the case of 5.5 years mean age, nearly all CFC-11 molecules are converted to the inorganic form and the remaining organic fraction gets very small. The mean transit times for all three distributions are calculated as the arithmetic mean or first moment of the respective distribution functions. The mean transit time of the organic fraction is described by the mean arrival time  $\Gamma^*$  (Plumb et al., 1999), that of an inert tracer by the mean age  $\Gamma$ . The inorganic fraction is described by a third time scale, which represents a release weighted transit time distribution. We suggest the term “release time distribution”,  $G^\#$ , for this transit time distribution with a first moment called the “mean release time”,  $\Gamma^\#$ . In the example given in Figure 1 the mean age  $\Gamma$  is 3 years, the mean arrival time  $\Gamma^*$  for the organic fraction is 1.75 years and the mean release time  $\Gamma^\#$



describing the inorganic fraction is 4.35 years. Inorganic chlorine thus lags the tropospheric time series more than expected from an inert tracer. Mean arrival time  $\Gamma^*$  and mean release time  $\Gamma^\#$  differ for each tracer depending on their chemical loss behavior, which is described by the fractional release factor  $f$ . A parameterization of the mean arrival time  $\Gamma^*$  for all the relevant chlorine and bromine species has been calculated as a function of lifetime and mean age (Plumb et al., 1999). The mean release time  $\Gamma^\#$  can be derived from  $\Gamma$ ,  $\Gamma^*$  and the fractional release factor (see section 3.1). As EESC is a proxy for inorganic halogen, we derive a new formulation of EESC which takes into account this interaction between chemistry and transport in an improved way.

### 3 Deriving a new formulation of EESC

The new mathematical formulation of EESC proposed here is derived based on the concept of how a trace gas of tropospheric origin with a temporal trend and chemical loss in the stratosphere propagates into the stratosphere. The organic source gases of chlorine and bromine are such gases. In order to derive the amount of inorganic chlorine or bromine that has been released from such an organic source gas at some point  $r$  in the stratosphere, three different functions must be considered, which are all functions of the transit time. We will denote transit time, i.e. the time a fluid element has spent in the stratosphere as  $t'$ , while the time itself will be denoted as  $t$ . First, the transit time distribution, i.e. how long it has taken for the individual fluid elements of this air parcel to travel from their entry point to the stratosphere to the location  $r$  in the stratosphere. ~~We will denote transit time, i.e. the time a fluid element has spent in the stratosphere as  $t'$ , while the time itself will be denoted as  $t$ .~~ Second, the temporal trend of the mixing ratios at the entry point has to be considered and, third, chemical loss during this transport. All three functions depend on the transit time  $t'$ . The integral over all possible transit times over these three functions will yield the remaining mixing ratio of the source gas. If we denote the time series at the entry point to the stratosphere as  $\chi_0(t - t')$ , the transit time distribution for air to reach some point  $r$  in the stratosphere as  $G(r, t')$  and the chemical loss term, which can be also be described by the factor  $(1 - f(t'))$ , where  $f(t')$  describes the fraction which has been lost, as  $e^{-\frac{t'}{\tau(t')}}$  we can describe the mixing ratio of the organic source gas at that point  $r$  in the stratosphere as

$$\chi_{strat}(r, t) = \int_0^\infty \chi_0(t - t') \cdot e^{-\frac{t'}{\tau(t')}} \cdot G(r, t') dt' = \int_0^\infty \chi_0(t - t') \cdot (1 - f(t')) \cdot G(r, t') dt' \quad (6)$$

The chemical loss term in (6) describes the loss by an exponential decay with lifetime  $\tau(t')$ , taking into account that the average lifetime is a function of the transit time. In a more general way, this loss can also be described by the factor  $(1 - f(t'))$ , where  $f(t')$  describes the fraction which has been lost. Consequently, as  $(1 - f(t'))$  is the remaining fraction of the organic source gas, the mixing ratio of inorganic chlorine released from the source gas would then be

$$\chi_{inorg, strat}(r, t) = \int_0^\infty \chi_0(t - t') \cdot f(t') \cdot G(r, t') dt' \quad (7)$$

For simplicity, we have assumed here that the source gas releases only one atom of inorganic halogen. Transport and mixing are described by the transit time distribution, also known as the age spectrum or Green's function  $G$ .  $G$  describes the probability of a certain transit time since entry into the stratosphere at the tropical tropopause, and thus describes both net mass transport and mixing.  $G$  is a function of transit time  $t'$  and the location in the stratosphere,  $r$ . The integral over the probability of all transit times must be equal to 1.

$$\int_0^{\infty} G(r, t') dt' = 1 \quad (8)$$

and the integral over all transit times weighted by their probability is the mean age of air  $\Gamma$  (Hall and Plumb, 1994).

$$\Gamma(r) = \int_0^{\infty} t' \cdot G(r, t') dt' \quad (9)$$

We now use the new transit time distribution  $G^\#$  that was introduced in section 2.  $G^\#$  is defined as the product of the transit time dependent fractional release factor  $f(t')$  and the age spectrum.

$$G^\#(r, t') \equiv f(t') \cdot G(r, t') \quad (10)$$

As  $G^\#$  is the product of the fractional release and the transit time distribution, it represents a release weighted transit time distribution. We will refer to this distribution as the release time distribution. Note that the integral over  $G^\#$  is only unity in case of complete loss of the organic fraction, i.e.  $f(t')$  is 1 for all transit times  $t'$ . In all cases, the integral must be less or equal to 1.

$$\int_0^{\infty} G^\#(r, t') dt' \leq 1 \quad (11)$$

We can, however, define a new, normalized release time distribution  $G_N^\#$ , by dividing  $G^\#$  through the integral of  $G^\#$  over all possible transit times

$$G_N^\#(r, t') \equiv \frac{G^\#(r, t')}{\int_0^{\infty} G^\#(r, t') dt'} = \frac{G^\#(r, t')}{\int_0^{\infty} f(t') \cdot G(r, t') dt'} \quad (12)$$

The integral over  $G_N^\#$  over all possible transit times is now unity

$$\int_0^{\infty} G_N^\#(r, t') dt' = 1 \quad (13)$$

The integral over all transit times weighted by the normalized release time distribution  $G_N^\#$  yields a "mean release time",  $\Gamma^\#$ , as also shown in Figures 1 and 2:

$$\int_0^{\infty} t' \cdot G_N^{\#}(r, t') dt' = \Gamma^{\#} \quad (14)$$

The integral in the denominator of (12) represents the first moment of the distribution of all fractional release factors, thus a mean fractional release factor, which is a function of the location  $r$  in the stratosphere (Ostermüller et al., 2017), but in contrast to  $f(t')$ , it is not a function of transit time anymore:

$$\int_0^{\infty} G^{\#}(r, t') dt' = \int_0^{\infty} f(t') \cdot G(r, t') dt' = \bar{f}(r) \quad (15)$$

Inserting (15) into (12) and solving for  $G^{\#}$  yields:

$$G^{\#}(r, t') = G_N^{\#}(r, t') \cdot \bar{f}(r) \quad (16)$$

5 Using the definition of  $G^{\#}$  (5) we can thus derive a relationship between  $G$  and  $G^{\#}$

$$G^{\#}(r, t') = G_N^{\#}(r, t') \cdot \bar{f}(r) = G(r, t') \cdot f(t') \quad (17)$$

The term  $f(t') \cdot G(r, t')$  in (7) can thus be replaced by  $G_N^{\#}(r, t') \cdot \bar{f}(r)$  to derive a new relationship for inorganic chlorine

$$\chi_{inorg, strat}(r, t) = \int_0^{\infty} \chi_0(t - t') \cdot \bar{f}(r) \cdot G_N^{\#}(r, t') dt' \quad (18)$$

In contrast to  $f(t')$ ,  $\bar{f}(r)$  is independent of  $t'$  and it can be extracted from the integral and (18) can be rewritten:

$$\chi_{inorg, strat}(r, t) = \bar{f}(r) \cdot \int_0^{\infty} \chi_0(t - t') \cdot G_N^{\#}(r, t') dt' \quad (19)$$

10 Instead of describing the mixing ratio of inorganic chlorine at some location  $r$  in the stratosphere, we can also describe it as a function of a certain mean age value  $\Gamma$ . This implies that at all locations  $r$  with the same mean release time, the release time distribution is the same. This assumption may not be valid everywhere, but as a mean age of 3 years is used for mid latitudes and of 5.5 years for high latitudes, we use this assumption only for air parcels under similar meteorological conditions (latitude bands). The release time distribution is then expressed as a function of mean release time  $\Gamma^{\#}$ . Equation (19) then becomes

$$\chi_{inorg, strat}(\Gamma, t) = \bar{f}(\Gamma) \cdot \int_0^{\infty} \chi_0(t - t') \cdot G_N^{\#}(\Gamma^{\#}, t') dt' \quad (20)$$

After multiplying the right hand side of (20) with the amount of halogen atoms released from a halocarbon ( $n_i$ ) and in case of bromine with the factor  $\alpha$  describing the relative efficiency of bromine and summing up over all halogen species  $i$  we arrive at

$$EESC_{new}(\Gamma, t) = \sum_{Cl} \left( n_i \bar{f}_i(\Gamma) \int_0^{\infty} \chi_{0,i}(t - t') G_{N,i}^{\#}(\Gamma^{\#}, t') dt' \right) + \alpha \sum_{Br} \left( n_i \bar{f}_i(\Gamma) \int_0^{\infty} \chi_{0,i}(t - t') G_{N,i}^{\#}(\Gamma^{\#}, t') dt' \right) \quad (21),$$

15 which is the new formulation we suggest for the calculation of EESC. This formulation is similar to the one used by Velders and Daniel (2014) and Newman et al. (2007) and also in the most recent WMO ozone assessment reports (Harris et al., 2014; L.J. Carpenter and S. Reimann et al., 2014; Montzka and Reimann et al., 2011), but differs in two aspects. First, chemical

loss is described by the new time independent fractional release factor  $\bar{f}$  (Ostermüller et al., 2017) and, second, instead of the age spectrum  $G$  with mean age  $\Gamma$  for an inert tracer the normalized release weighted distribution function  $G_N^\#$  with the corresponding mean release time  $\Gamma^\#$  for a chemically active specie is used.

In order to apply this new formulation of EESC, the normalized release time distribution  $G_N^\#$  and the time independent fractional release factors  $\bar{f}$  (Ostermüller et al., 2017) for all relevant chlorine and bromine species need to be known. To our knowledge, so far only mean arrival times time  $\Gamma^*$  for most species are available from the literature (Plumb et al., 1999). In the following we therefore show how the first moment of  $G_N^\#$  (the mean release time  $\Gamma^\#$ ) and  $\bar{f}$  can be derived from the information available.

### 10 3.1. Deriving the mean release time.

The arrival time distribution  $G^*$  (Plumb et al., 1999) and the release time distribution  $G^\#$  used here are closely linked. By combining equation (4) and (5) it is easily shown that the sum of  $G^*$  and  $G^\#$  is the age spectrum  $G$ :

$$G^\#(r, t') + G^*(r, t') = f(t') \cdot G(r, t') + (1 - f(t')) \cdot G(r, t') = G(r, t'). \quad (22)$$

In a similar way as for  $G^\#$ , a normalized arrival time distribution  $G_N^*$  has been defined (Ostermüller et al., 2017):

$$G_N^*(r, t') = \frac{G^*(r, t')}{(1 - \bar{f}(r))}. \quad (23)$$

In a similar way as for the mean release time, a mean arrival time  $\Gamma^*$  (Plumb et al., 1999) can be derived as the first moment of the arrival time distribution (4). In the arrival time distribution the age spectrum is not weighted with the release term  $f(t')$  (as is the case for  $G^\#$ ), but rather with the loss term  $(1 - f(t'))$ ,

$$\int_0^\infty t' \cdot G_N^*(r, t') dt' = \Gamma^* \quad (24)$$

$\Gamma^*$  has been calculated in a 2D model and is available for all relevant halocarbons based upon a parameterization (Plumb et al., 1999) as function of  $\Gamma$  and lifetime  $\tau$ .  $\Gamma^\#$ , which is needed to calculate the release of inorganic halogen from a halocarbon can be derived from the knowledge of  $\Gamma$ ,  $\Gamma^*$  and fractional release factors. To derive  $\Gamma^\#$  we start with the relationship between  $G^\#$  and  $G^*$  (22).

Multiplying (22) with  $t'$  and integrating over all possible transit times yields

$$\int_0^\infty t' \cdot G(r, t') dt' = \int_0^\infty t' \cdot G^*(r, t') dt' + \int_0^\infty t' \cdot G^\#(r, t') dt'. \quad (25)$$

Replacing  $G^*$  and  $G^\#$  with their normalized distributions  $G_N^*$  (23) and  $G_N^\#$  (16) yields

$$\int_0^\infty t' \cdot G(r, t') dt' = \int_0^\infty t' \cdot (1 - \bar{f}(r)) \cdot G_N^*(r, t') dt' + \int_0^\infty t' \cdot \bar{f}(r) \cdot G_N^\#(r, t') dt'. \quad (26)$$

Extracting the transit time independent fractional release factors (Ostermüller et al., 2017) from the integrals yields

$$\int_0^{\infty} t' \cdot G(r, t') dt' = (1 - \bar{f}(r)) \cdot \int_0^{\infty} t' \cdot G_N^*(r, t') dt' + \bar{f}(r) \cdot \int_0^{\infty} t' \cdot G_N^{\#}(r, t') dt' \quad (27)$$

All the integrals in (27) can be solved as they are the first moments of the respective distribution functions, thus mean age  $\Gamma$  (9), mean arrival time  $\Gamma^*$  (24) and mean release time  $\Gamma^{\#}$  (14). (27) thus becomes

$$\Gamma = (1 - \bar{f}(r)) \cdot \Gamma^* + \bar{f}(r) \cdot \Gamma^{\#} \quad (28)$$

Again we express  $\bar{f}$  as a function of mean age  $\Gamma$  instead of location  $r$  and then rearrange (28) to give an equation to calculate

5  $\Gamma^{\#}$ :

$$\Gamma^{\#} = \frac{\Gamma - (1 - \bar{f}(\Gamma)) \cdot \Gamma^*}{\bar{f}(\Gamma)} \quad (29)$$

$\Gamma^{\#}$ , which is the first moment of  $G_N^{\#}$ , can thus be derived based on the mean fractional release factor, mean age and  $\Gamma^*$  for each compound. In the next section we now derive a formulation to calculate the mean fractional release factors  $\bar{f}(\Gamma)$  from available fractional release factors.

### 3.2. Recalculating FRF values to yield time-independent mean FRF values $\bar{f}$ .

10 For the calculation of mean release time  $\Gamma^{\#}$ , and also in the new formulation for EESC (21) the time independent mean fractional release factors  $\bar{f}$  as derived by (Ostermüller et al., 2017) are needed.

$$\bar{f}(\Gamma) = \frac{\int_0^{\infty} \chi_0(t - t') \cdot G_N^*(\Gamma^*, t') dt' - \chi_{strat}(\Gamma, t)}{\int_0^{\infty} \chi_0(t - t') \cdot G_N^*(\Gamma^*, t') dt'} \quad (30)$$

The fractional release factors from the most recent WMO reports are largely based on observations from the time period 1996 to 2000 (Newman et al., 2007) and were derived using

$$f(\Gamma) = \frac{\int_0^{\infty} \chi_0(t - t') \cdot G(\Gamma, t') dt' - \chi_{strat}(\Gamma, t)}{\int_0^{\infty} \chi_0(t - t') \cdot G(\Gamma, t') dt'} \quad (31)$$

In this formulation, the age spectrum for an inert tracer is used, which does not include chemical loss. Solving (31) for

15  $\chi_{strat}(\Gamma, t)$  and inserting this into (30) yields

$$\bar{f}(\Gamma) = \frac{\int_0^{\infty} \chi_0(t - t') \cdot G_N^*(\Gamma^*, t') dt' - (1 - f(\Gamma)) \cdot \int_0^{\infty} \chi_0(t - t') \cdot G(\Gamma, t') dt'}{\int_0^{\infty} \chi_0(t - t') \cdot G_N^*(\Gamma^*, t') dt'} \quad (32)$$

Equation (32) allows to convert fractional release factors  $f$  calculated according to Newman et al. (2007) to time-independent values  $\bar{f}$  according to Ostermüller et al. (2017). We derived  $\bar{f}$  for every month of the period 1996 to 2000 from  $f$  and then

took the median of these values. The new  $\bar{f}$  values and the spread of  $\bar{f}$  values derived by the conversion during the different months of this period are shown in Tables 1 and 2. The spread is mostly very small, as temporal trends during this period were small for many species. For the same reason, the  $\bar{f}$  values derived in this way mostly do not differ very strongly from the  $f$  values, as  $f$  and  $\bar{f}$  only differ due to tropospheric trends.

5 The fractional release factors used for the reference calculation presented in section 4 are those used by Velders and Daniel (2014), modified using equation (32) to be consistent with the new formulation given by Ostermüller et al. (2017). The mean release time  $\Gamma^*$  has been calculated according to the parameterization of Plumb et al. (1999) also using their model lifetimes. Plumb et al. (1999) used a 2D model for their study. Despite this, the stratospheric lifetimes derived from the model are in overall good agreement to more recent model studies (Chipperfield et al., 2013). The sensitivity of the parameterization  
10 between mean release time and mean age to the stratospheric lifetimes is further discussed in section 4. For CFCs 114 and 115 which are not included in Plumb et al. (1999) we used stratospheric lifetimes from Ko et al. (2013), while the stratospheric lifetimes for HCFC-142b and the halons halon-1301, halon-1202 and halon-2402, which are also not included ~~by~~ Plumb et al. (1999), are taken from Chipperfield et al. (2013). For those species included in Plumb et al. (1999) we used the species specific fit parameters, while for other species we used the averaged fit parameters reported in Plumb et al. (1999). The values  
15 for lifetimes used in the calculations and for both mean arrival time and mean release time as well as the mean fractional release factors  $\bar{f}$  are given for all species used in this calculation in Tables 1 and 2.

#### 4. Temporal evolution of EESC and implications for recovery to 1980 benchmark values.

The new formulation of EESC (21) uses a loss weighted transit time distribution, the release time distribution, and different  
20 fractional release factors from those used in the ~~classical~~-formulation (Newman et al., 2007). The new fractional release factors are based on the formulation suggested by Ostermüller et al. (2017) and have been derived from available fractional release factors (see section 3.2). No method to calculate the release time distribution is available so far. Both the mean release time  $\Gamma^\#$  (first moment of the distribution) and the shape of the release time distribution  $G_N^\#$  need to be known in order to use this distribution for the calculation of the propagation of tropospheric trends into the stratosphere. The age spectrum for an inert  
25 tracer,  $G$ , is commonly described by an inverse Gaussian function with a parameterization of the width as function of mean age (Hall and Plumb, 1994; Schauffler et al., 2003; Newman et al., 2007). As no such parameterization has yet been established for  $G_N^\#$  we have assumed that the general shape of  $G_N^\#$  is similar to that of  $G$ , with  $\Gamma^\#$  instead of  $\Gamma$  as mean value. The sensitivity of our calculations to these assumptions is discussed in section 4.2.

#### 4.1. Comparison of different EESC formulations

As already mentioned, new time-independent fractional release factors and the release time distribution are needed for our new formulation of EESC. The release time distribution is approximated assuming the form of an inverse Gaussian with a species specific first moment  $\Gamma^{\#}$  and a width of  $\lambda = \frac{\Delta^{\#2}}{\Gamma^{\#}} = 0.7 \text{ years}$ .

- 5 The new time-independent fractional release factors are based on the concept of arrival time distribution (Plumb et al., 1999). Ostermüller et al. (2017) showed that using this concept, fractional release factors can be calculated, which are independent of time, as long as stratospheric transport or photochemistry remain unchanged. More specifically, these fractional release factors are independent of the tropospheric trend of the respective species. We have recalculated fractional release factors used in the most recent ozone assessment report (Harris et al., 2014) to be consistent with the new formulation of fractional release. The
- 10 fractional release factors commonly used are largely based on observations (Newman et al., 2007), except for the hydrochlorofluorocarbons HCFC-141b and HCFC-142b (Daniel et al., 1995) (see Tables 1 and 2). Other observation based fractional release factors have been presented by Laube et al. (2013). The uncertainty due to the use of different fractional release factors, different emissions and different lifetimes have been discussed in details by Velders and Daniel (2014). Here, we focus on the uncertainties due to the suggested new formulation for the calculation of EESC. Using these new FRF values
- 15 and the mean arrival time  $\Gamma^*$  based on the available parameterization (Plumb et al., 1999), we have calculated values for  $\Gamma^{\#}$  for all relevant chlorine and bromine containing source gases (see Tables 1 and 2).

- Figures 3 and 4 show the calculation according to (21) using the new time independent FRF values for 3 and 5.5 years of mean age, respectively, and compare it with the calculation applying formulation (3) using the FRF values of the ozone assessment reports (Harris et al., 2014). All values given here are mole fractions given in ppt, which is equivalent to pmol/mol. The values
- 20 are also summarized in Tables A1 and A2. The tropospheric time series and the future projection used for this calculation are based on Velders and Daniel (2014), where updated lifetimes (Ko et al., 2013) and assumptions on future emissions have been used as basis for the projection of tropospheric time series. For a mean age of air of 3 years, as used for mid-latitudes, there are significant differences between the two methods used for calculation of EESC (see Figure 3). In the case of our new formulation, there is a longer time lag between the troposphere and the arrival of the inorganic halogen in the stratosphere.
- 25 The tropospheric halogen loading was increasing strongly during the time before 1980, and therefore EESC at that time was dominated by air masses that had a lower halogen content. As a consequence, we calculate 1980 EESC levels in the mid latitude lower stratosphere which are about 90 ppt lower (see Table 3 for details) than using the EESC formulation according to Newman et al. (2007). During the recovery phase of stratospheric halogen loading, temporal trends of halogenated source gases in the troposphere will be negative; EESC will thus be dominated by air masses with higher chlorine content and is
- 30 higher in our new formulation. In combination with the lower level of EESC, which must be attained for recovery, a significantly later recovery date is calculated. According to our calculation, mid latitude lower stratospheric EESC levels will return to 1980 values in 2060 only, which is more than 10 years later than the recovery date of 2049 calculated using the current method (Velders and Daniel, 2014).

For polar winter conditions (5.5 years of mean age) shown in Figure 4, the recovery date calculated here is 2077, relative to a value of 2076 derived based on the currently used method using the same scenario (Velders and Daniel, 2014). The reason that only a very minor change is calculated for polar winter conditions is that under these conditions nearly all source gases are converted to their inorganic form and the differences between the age spectrum and the release time distribution become very small.

#### 4.2. Sensitivity discussion and tests

As mentioned above, we will concentrate on the sensitivity of the new EESC method on the limited knowledge on the new release time distribution  $G_N^\#$  and on the new fractional release factors (Ostermüller et al., 2017) used here. We have therefore performed sensitivity calculations to evaluate the sensitivity of our results on the changed fractional release factors and on the uncertainty in the knowledge of the release time distribution  $G_N^\#$ .

##### Sensitivity to new fractional release factors

To evaluate the changes due to the changes in fractional release factors, we use our new release time distribution  $G_N^\#$  but use the same fractional release factors as in previous studies (Velders and Daniel, 2014; Harris et al., 2014). The comparison for a mean age of 3 years is shown in Figure 5 and Table 3 (bottom row). The estimated recovery year is 2058, instead of 2060 using our new fractional release factors. The change in maximum EESC is also small with a value of 1909 using our new fractional release factors and 1895 using the fractional release factors as in Velders and Daniel (2014). For 5.5 years of mean age (not shown), the same recovery date is calculated (2077) with about 20 ppt lower EESC during the maximum using the unmodified fractional release factors from Velders and Daniel (2014). The new time-independent fractional release factors are derived from the fractional release factors presented by Newman et al. (2007) as described in the Appendix. For this, a correction needs to be applied (see Appendix) based on the year of the measurements from which the fractional release factors have been derived. The fractional release factors used by Newman et al. (2007) were derived from the measurements taken during the STRAT campaign (1996), the POLARIS campaign (1997) and from the SOLVE ~~campaign (1999-2000)~~ (Schauffler et al., 2003)- ~~campaign (1999-2000)~~. We converted the fractional release factors assuming that they were taken during the time period 1996-2000, with the exception of HCFC-s 141b and 142b, where we used the values given in Velders and Daniel (2014). We performed the conversion for every month of this period. The median of all values was taken as the best estimate for the new time-independent fractional release factors. We also derived a variability which was below 1% for most species. This variability is also presented in Tables 1 and 2 together with the new fractional release factors. As a sensitivity test, we performed the EESC calculation by shifting all fractional release values up or downwards by  $1\sigma$ . Varying the new fractional release factors within this uncertainty range resulted in an upward (increased fractional release) or downward (decreased fractional release) shift of EESC by about 14 ppt during the maximum of EESC for the 3 year mean age calculation. The



changes in the calculated recovery years were less than 0.2 years. For 5.5 years of mean age, the variation of the fractional release factors lead to even smaller changes. Overall, we conclude that the changes in the fractional release factors are rather small in comparison to the overall changes. ~~Using the fractional release factors recalculated according to the method of Ostermüller et al. (2017) leads to change of 2 years in the calculated recovery date for mid latitudes; varying the recalculated fractional released factors within their uncertainties only led to very small changes.~~ For polar winter conditions, the calculated changes were in all cases very small. The significant differences in recovery dates for the mid latitude lower stratosphere presented above are thus mainly due to the new release time distribution  $G_N^\#$ .

### Sensitivity to the shape of the new release time distribution $G_N^\#$

The new release time distribution  $G_N^\#$  has not been calculated from models to our knowledge. We therefore have to make assumptions on the shape and the width of  $G_N^\#$ . In the calculation presented above, we have assumed that the shape is similar as for  $G$ , i.e. an inverse Gaussian function. We then made the assumption that the width can be described in a similar way as a function of the first moment, i.e. using a constant factor  $\lambda = \frac{\Delta^2}{\Gamma} = \frac{\Delta^{\#2}}{\Gamma^\#} = 0.7 \text{ years}$  (Hall and Plumb, 1994; Engel et al., 2002). In order to test the sensitivity of our results to these assumptions, we have varied this parameter between values of 0 and 2 years in the calculation of  $G_N^\#$ , while retaining the first moment, i.e.  $\Gamma^\#$ . The extreme case of 0 would mean that the release time distribution  $G_N^\#$  collapses to one single transit time or lag time, i.e.  $\Gamma^\#$  and that no mixing occurs during the transport in the stratosphere. In this case, stratospheric inorganic chlorine is simply derived by time shifting the tropospheric time series of a source gas by the time lag  $\Gamma^\#$  and multiplying it with the fractional release factor.

The scenarios with  $\lambda$  equal to 0 years (no mixing case), 0.7 years (reference case) and 2 years (strong mixing case) are compared in Figure 6 and 7 for mean age values of 3 and 5.5 years. It is obvious, that the calculation of the recovery year is not very sensitive to the width of the release time distribution  $G_N^\#$ . In the case of the pure lag time calculation assuming no mixing ( $\lambda = 0 \text{ years}$ ) recovery is about 1 year later for both 3 and 5.5 years of mean age compared to the reference case ( $\lambda = 0.7 \text{ years}$ ). In the case of a very wide spectrum assuming strong mixing ( $\lambda = 2 \text{ years}$ ), recovery is expected about 1.5 years earlier at the mean age level of 3 years and 2.5 years earlier at the mean age level of 5.5 years.

The overall range of the calculated recovery dates is 2.2 years in the case of 3 years mean age and 3.8 years in the case of 5.5 years of mean age. The recovery dates and the maximum values of EESC calculated under the different assumptions are compared in Table 4. This rather small dependence on the width of the applied transit time distribution even for the assumption of extreme cases is due to two factors. Firstly, the deviation of tropospheric trends from linearity in the years prior to the reference year of 1980 and during the recovery phase after 2030 are rather small, in which case the propagation values becomes independent of ~~the~~ the shape of the distribution (Hall and Plumb, 1994) and only depend on the first moment, i.e. the mean release time  $\Gamma^\#$ . Secondly, during the period of the maximum EESC, tropospheric trends are rather small overall and thus the trend correction becomes rather small. Therefore maximum differences are below 50 ppt both for 1980 values and for the maximum

EESC values for 3 years of mean age and 73 ppt for 5.5 years of mean age. The largest dependence on the parameterization is thus derived for 5.5. years of mean age during the maximum of EESC, as tropospheric data from a large time period need to be taken into account here and there is significant non-linearity in the trends. In all cases, the lag time calculation yields higher EESC during the maximum, as would be expected.

#### 5 Sensitivity to the mean release time derived from mean age and stratospheric lifetime.

Another source of uncertainty is, that the stratospheric lifetime of the individual compounds needs to be known in order to calculate the mean arrival time  $\Gamma^*$  from which the mean release time  $\Gamma^\#$  is derived (29). We tested the sensitivity of our calculation to that by systematically increasing all lifetimes by 20% or decreasing them by 20% (see Table 5) in the parameterization given by (Plumb et al., 1999). This results in different mean arrival time  $\Gamma^*$  and mean release time  $\Gamma^\#$ . Even if such rather large changes would go in the same direction for all species, the impact on our calculations is rather small. For 3 years of mean age, the calculated maximum in EESC varied by only 6 ppt and the calculated recovery date varied by 2.5 years. For 5.5 years of mean age the effect is even smaller with variation in maximum EESC of less than 1 ppt and a variation of less than half a year in the recovery date. This very small sensitivity at 5.5 years of mean age is due to most fractional release factors being close to 1 under these conditions. The reason for this small effect is that next to the parameterization of the mean arrival time, the fractional release factor determines the mean release time. Therefore, the sensitivity of the mean arrival time to the assumed lifetime in our calculation is quite low. Consequently, the influence on the derived EESC is also rather small. Despite this rather low sensitivity, it should be noted that the parameterization is derived from a 2D model. The relationship between mean age, age spectrum and chemical loss should be explored in state-of-the-art 3 D models, which have a better representation of stratospheric transport processes.

#### 20 4.3. Comparison of EESC formulations with model calculations of inorganic halogen loading.

In order to evaluate our new formulation of EESC we have compared the results of our calculations with the inorganic halogen loading calculated from two comprehensive three-dimensional atmospheric chemistry models. Due to expected long-term changes in mean age on a given pressure level associated with the simulated changes in the Brewer-Dobson circulation (e.g. Butchart, 2014; Austin and Li, 2006), changes in fractional release factors on mean age levels are also observed in free running model calculations (Douglass et al., 2008; Li et al., 2012b). We have therefore compared our new formulation of EESC to model calculations with changing and with annually repeating ('fixed') dynamics. To compare the new formulation with the formulation by (Newman et al., 2007), we used a model simulation from the TOMCAT model (Chipperfield et al., 2017), which was driven by a repeated meteorology, in this case for the year 1980. Effects due to changing dynamics, which are not included in the concept of EESC, will thus not impact this calculation, making it an ideal test bed for comparison of the two formulations. For long term changes, we have used model results from the EMAC model (Jöckel et al., 2016), which includes

5 expected changes in stratospheric transport. As in general the relationship between mean age and  $Cl_v$  is very different for  
different models (Waugh et al., 2007), a direct comparison between EESC and ESC (Equivalent stratospheric Chlorine,  
calculated from model  $Cl_v$  and  $Br_v$  using the same sensitivity parameters for bromine as with EESC) is not meaningful, as  
differences may be due to different fractional release factors between models and observations. Instead we have used fractional  
10 release values derived from the models for 3 years of mean age and used these for the calculation of EESC using the  
formulations by Newman et al. (2007) and from this work. Fractional release factors were calculated from the model data  
using the methods of Newman et al. (2007) and Ostermüller et al. (2017) for this work. The fractional release factors were  
calculated for the year 2000, in order to be consistent with the observation based fractional release factors, which were derived  
mainly for the year 2000. To the EESC calculated in this way we added simulated inorganic chlorine and bromine at the  
15 tropical tropopause, which we propagated as an inert tracer. VSLS (very short lived substances) were treated in a similar way,  
using their tropical tropopause values as input, as loss in the troposphere cannot be neglected for these species. As no global  
stratospheric lifetimes are available for these species, it is not possible to apply the new formulation. Therefore, the VSLS  
were treated using the method of Newman et al. (2007). The differences are negligible, as the VSLS have rather slow long-  
term trends and both methods yield nearly identical results, as also discussed in Ostermüller et al. (2017). As some loss of  
20  $CH_3Cl$  and  $CH_3Br$  occurs during the transport in the troposphere to the tropical tropopause, we have also used the time series  
of these two gases at the tropical tropopause rather than at the surface in these calculations.

### **Comparison to fixed dynamics model calculations**

For comparison of the two formulations to model calculations with fixed dynamics, we used a TOMCAT model run  
(Chipperfield et al., 2017), which is driven by repeated 1980 meteorology. This model run is available from 1960 through to  
20 2016. The fractional release factors derived from the model for the northern hemisphere are significantly higher as function of  
mean age than the observed fractional release values. Southern hemispheric fractional release values for 3 years of mean age  
showed better agreement with observation derived fractional release factors (Newman et al., 2007). For this reason we  
compared simulated ESC from the southern hemisphere with EESC calculated using our new formulation and the formulation  
by (Newman et al., 2007), in both cases using fractional release values derived for the year 2000 model results.  
25 Figure 8 shows the comparison between the modelled ESC and EESC for a mean age of 3 years calculated as described above,  
including all bromine and chlorine species included in the model and also including inorganic chlorine and bromine entering  
the stratosphere. As the differences between the two formulations of EESC are most pronounced for 3 years of mean age, we  
show this comparison for 3 years of mean age only. A much better agreement is observed when applying the new formulation  
than using the formulation by Newman et al. (2007), due to the improved treatment of the combined influence of transport and  
30 mixing on chemical loss. Remaining discrepancies between model ESC and EESC are most probably due to an imperfect  
parameterization of the loss time distribution  $G^\#$ .

## Comparison to model calculations with varying dynamics

Under changing stratospheric dynamics (e.g. Butchart, 2014), it is expected that fractional release factors at a given mean age level will change (Douglass et al., 2008; Li et al., 2012b; Ostermüller et al., 2017). Therefore, the inorganic halogen loading as a function of mean age would be expected to change even if all source gases remained constant in time. Under such conditions, EESC is not expected to follow ESC on a given mean age level. To estimate the validity of EESC as a proxy for inorganic halogen loading of the stratosphere, we have compared our new formulation to a free running chemistry climate model simulation. We used data from the EMAC model simulation RC2-base-04 from the ESCiMo project for this (Jöckel et al., 2016). This simulation covers the 1950–2100 time frame with simulated sea surface temperatures and sea ice contents. As described above, we again calculated fractional release factors from the model in order to have results which are internally consistent. Northern Hemisphere fractional release factors for the year 2000 are in good agreement with observation based fractional release factors (Newman et al., 2007; Laube et al., 2013) and therefore we used northern hemispheric data for this comparison. In addition to comparing ESC on a fixed mean age level we also compared ESC on a fixed pressure level to our new formulation of EESC. A similar comparison has been presented in Shepherd et al. (2014), who compared model ESC on a fixed pressure level to EESC on fixed mean age level (using the formulation of Newman et al. (2007)), showing good agreement. Figure 9 compares the time evolution of EESC for 3 years of mean age with model ESC at the 60 hPa level (corresponding to 3 years of mean age in the year 2000) and model ESC at 3 years of mean age. The year of 2000 and the corresponding level of 60 hPa was chosen, as we also evaluated fractional release factors in the year 2000 of the model run. As expected, ESC at 3 years of mean age deviates systematically from EESC, especially in the future when fractional release evaluated on a mean age surface changes significantly in the model. The agreement with ESC on a fixed pressure level is however much better. In this comparison EESC at 3 years of mean age would slightly overestimate ESC on a pressure level in the future and significantly underestimate ESC on a mean age level. The exact magnitude of changes in stratospheric dynamics is highly uncertain and also we expect ozone to follow a pressure surface rather than a mean age surface in the future. We therefore conclude that EESC is a reasonable proxy for the effect of halogen loading on stratospheric ozone, given the overall high uncertainties associated to the future evolution of stratospheric dynamics.

## **5. Conclusions and outlook**

We have shown that for the calculation of the propagation of chlorine and bromine source gases with photochemical loss, different transit time distributions must be used to calculate the amount of organic ~~-, respective or~~ inorganic chlorine present at a given mean age level. First, treating the propagation of these tracers with the age spectrum for an inert tracer leads to fractional release values, which show a strong temporal variability in case of large tropospheric trends of the respective gas (Ostermüller et al., 2017). ~~Therefore, time independent~~ Fractional release factors, which are independent of the tropospheric trends (Ostermüller et al., 2017), must be used to correctly describe the fraction that has been transferred from the organic source gas

to the inorganic form and can then influence ozone chemistry. Secondly, changes in tropospheric mixing ratios lead to changes in stratospheric inorganic halogen with a time delay that is longer than the mean age, which describes the propagation of an inert tracer. This can be described by a modified transit time distribution, in which the transit times from the classical age spectrum are weighted with the chemical loss during this transport time. We suggest the term “release time distribution” for this modified tracer specific transit time distribution.

We developed a new formulation of EESC, which uses the release time distribution and time-independent fractional release factors calculated by the method of Ostermüller et al. (2017). This approach more accurately represents the amount of Cl<sub>y</sub> and Br<sub>y</sub> in the stratosphere from tropospheric source gas concentrations and fractional release factors, as shown in comparison to a model calculation with annually repeating dynamics. We have shown that the long-term evolution of equivalent stratospheric chlorine (ESC, i.e. inorganic chlorine and bromine, the latter weighted in a similar way as in EESC to reflect the higher efficiency of bromine to ozone depletion) in the model deviates substantially from our calculation of EESC in a long-term model calculation with varying dynamics. However, we have also shown that the new formulation of EESC is a reasonable proxy for the evolution of inorganic halogen loading on a given pressure level. We therefore conclude that EESC is a reasonable proxy for future halogen impact on ozone. We suggest our new method should be adopted~~We suggest that this new method~~ to calculate EESC ~~should be and to adopted to~~ estimate the time of recovery of inorganic halogen to 1980 values under otherwise unchanged conditions. This will lead to a delay of about 10 years in ~~expected-calculated chlorine-EESC~~ recovery in the lower mid-latitude stratosphere (mean age of 3 years) compared to the formulation currently used (Velders and Daniel, 2014; Newman et al., 2007), and also applied in the WMO ozone assessment reports (Harris et al., 2014; L.J. Carpenter and S. Reimann et al., 2014). If all other factors ~~are-were~~ unchanged, in particular stratospheric dynamics, the recovery of ~~the mid-latitude lower stratospheric ozone~~ to unperturbed values of chlorine and bromine is-would thus also expected to take about 10 years longer than previously estimated using EESC based on the formulation by Newman et al. (2007). As current climate models consistently predict an acceleration in the Brewer-Dobson circulation (Butchart, 2014), this will have an impact on the temporal evolution of inorganic halogen loading of the stratosphere. These expected changes in the Brewer-Dobson circulation would result in an earlier recovery of ozone at mid- and high latitudes (Eyring et al., 2010). These changes are not included in the concept of Shepherd et al. (2014)EESC. However, we have shown that EESC is a reasonable proxy for ESC when ESC is evaluated at constant pressure level. (Eyring et al., 2010)In addition to this, increases in the concentrations of N<sub>2</sub>O and short-lived chlorine-containing halocarbons may further influence retard the recovery of the ozone layer, possibly leading to a later recovery (Hossaini et al., 2015b; Hossaini et al., 2015a; Chipperfield, 2009). The changes due to application of ~~this-our~~ new method for 5.5 years of mean age (representative of polar winter conditions) are rather small compared to the formulation suggested by Newman et al. (2007), as nearly all halogen is released under these conditions and the difference between age spectrum and release time distribution becomes small.

The two changes relative to the currently used formulation for EESC are the use of new time-independent fractional release factors and of the release time distribution. We have shown that the new time-independent fractional release factors do not differ very much from the fractional release factors currently used (Harris et al., 2014; Velders and Daniel, 2014), as they were

derived during a period of rather small tropospheric trends for many species. Consequently, the projected EESC recovery dates vary by 2 years or less depending on which fractional release factors are used. We have also shown that the calculation of the recovery date shows some sensitivity to the assumed width of the release time distribution, with variations of about 2 years for the mid-latitude calculations and 3.5 years for the high latitude case. Varying the stratospheric lifetimes assumed for the calculation of the loss weighted transit distribution  $G^{\#}$  a similar influence on the projected recovery dates for 3 years mean age (mid-latitude conditions) and virtually no effect for 5.5 years (high-latitude conditions) is derived. In general, the maximum EESC level is more sensitive to variations in the assumed width of the release time distribution than to the stratospheric lifetimes assumed in the calculation of the mean release time. The strongest dependence on the assumed width is observed during the maximum of EESC levels, especially for high-polar winter conditions, as tropospheric trends were strongly non-linear during that time. A more realistic description of the shape of the release time distribution would improve especially the prediction of EESC during its maximum. Age spectra for inert tracers in models for atmosphere and ocean have been derived from pulse experiments using tracers without chemical loss (Haine et al., 2008; Li et al., 2012c; Li et al., 2012a; Ploeger and Birner, 2016). For the derivation of the release time distribution and improved information on mean release time such pulse experiments for tracers with chemical loss should be performed. Such calculations are only available based on a rather old 2D model (Plumb et al., 1999) and should be repeated with state-of-the-art models. These release time distributions will be specific for each tracer, but should generally be similar for species with similar lifetimes and similar loss processes. ~~Using such an improved release time distributions a comparison of inorganic halogen levels from full model calculations with the new EESC method should be performed. As we have shown good agreement with model calculations using the parameterization suggested here, we do not expect a re-evaluation of the loss weighted age spectra to lead to large changes. As the suggested reformulation of EESC does not affect the principal behavior of the temporal evolution of EESC, we do not expect this reformulation to lead to substantial changes, which could impact the changes of studies using EESC except for those which have used EESC to project EESC recovery.~~ For studies using EESC as a proxy for the halogen loading, e.g. in comparison to ozone time series, the new formulation of EESC suggested here should nevertheless be used, as the timing of the recovery especially for mid latitudes is significantly different than in previous estimates.

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*Data availability:* The work is based on the scenario developed by (Velders and Daniel, 2014). We have used the scenario “All\_parameters\_SPARC2013\_mostlikely\_mc.dat” given in the Appendix to that paper and available as supplement. For data availability of the EMAC simulation results we refer to Jöckel et al., (2016). The TOMCAT model results are available by emailing Martyn Chipperfield—.

30 *Author contribution:* . Andreas Engel has performed all the most of the calculations in the manuscript, has written the manuscript and has developed the ideas presented in the manuscript together with the ~~co-authors~~ Jennifer Ostermüller and Harald Bönisch in the frame of many open discussions. Jennifer Ostermüller and Harald Bönisch have both participated in the discussion and preparation of the manuscript. Jennifer Ostermüller has performed the calculations for the comparison with model data (section 4.3.). Sandip Dhomse and Martyn Chipperfield have provided TOMCAT model results. Patrick Jöckel is PI of the ESCiMo

project, conducted the EMAC model simulations and provided the corresponding data. All authors were involved in the final revision of the manuscript.

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**Table 1:** Mean arrival time  $\Gamma^*$ , mean release time  $\Gamma^\#$ ,  $f$  and  $\bar{f}$  for all relevant long lived chlorine and bromine species for a mean age of 3 years. The time independent fractional release factors were derived using equation (32). The measurements from which the original fractional release factors were derived are from the period 1996-2000. We calculated the conversion using eq. (32) for every month of this period for all species, except for HCFC-141b and 142b (see text). The  $1\sigma$  variability of the converted fractional release factors is also shown.

	Stratospheric lifetime used for $\Gamma^*$ [years]	mean arrival time $\Gamma^*$ [years]	mean release time $\Gamma^\#$ [years]	fractional release factor $f$	time independent fractional release factor $\bar{f}$	$1\sigma$ variability of recalculated $\bar{f}$ in %
		Mean age $\Gamma$ of 3 years				
CFC-11	55.7 <sup>a</sup>	1.5	4.7	0.47 <sup>d</sup>	0.47	0.23
CFC-12	102 <sup>a</sup>	2.0	6.2	0.23 <sup>d</sup>	0.24	0.22
CFC-113	84.5 <sup>a</sup>	1.9	5.7	0.29 <sup>d</sup>	0.30	0.69
CFC-114	189 <sup>b</sup>	2.2	8.7	0.12 <sup>d</sup>	0.13	0.11
CFC-115	1020 <sup>b</sup>	2.5	12.1	0.04 <sup>d</sup>	0.07	0.45
CCl <sub>4</sub>	48.4 <sup>a</sup>	1.4	4.3	0.56 <sup>d</sup>	0.56	0.08
CH <sub>3</sub> CCl <sub>3</sub>	48.8 <sup>a</sup>	1.4	4.1	0.67 <sup>d</sup>	0.61	1.93
HCFC-22	217 <sup>a</sup>	2.6	5.6	0.13 <sup>d</sup>	0.15	0.09
HCFC-141b	73 <sup>a</sup>	1.8	5.4	0.34 <sup>e</sup>	0.34	n.a. <sup>f</sup>
HCFC-142b	212 <sup>c</sup>	2.2	6.8	0.17 <sup>e</sup>	0.17	n.a. <sup>f</sup>
halon-1211	39.5 <sup>a</sup>	1.3	4.0	0.62 <sup>d</sup>	0.65	0.4
halon-1202	15.3 <sup>c</sup>	0.5	5.0	0.62 <sup>d</sup>	0.67	2.79
halon-1301	70.8 <sup>c</sup>	1.7	6.0	0.28 <sup>d</sup>	0.32	1.34
halon-2402	33.8 <sup>c</sup>	1.2	4.0	0.65 <sup>d</sup>	0.66	0.55
CH <sub>3</sub> Br	40.2 <sup>a</sup>	1.3	4.1	0.60 <sup>d</sup>	0.60	0.11
CH <sub>3</sub> Cl	63.7 <sup>a</sup>	1.9	4.4	0.44 <sup>d</sup>	0.44	0 <sup>g</sup>

<sup>a</sup> stratospheric lifetime from the 2D model used ~~in~~ by Plumb et al. (1999)

<sup>b</sup> total atmospheric lifetime taken from recommendations in SPARC lifetime assessment (Ko et al., 2013), as tropospheric loss is negligible.

10 <sup>c</sup> stratospheric lifetime taken from modelling work for SPARC lifetime assessment (Chipperfield et al., 2013).

<sup>d</sup> fractional release values based on Newman et al. (2007)

<sup>e</sup> fractional release value based on parameterization given in footnote to table 2 in Velders and Daniel (2014).

<sup>f</sup> not applicable, as the fractional release values have not been recalculated.

<sup>g</sup> no variability was derived as there is no trend in the tropospheric reference data applied.

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**Table 2:** Mean arrival time  $\Gamma^*$ , mean release time  $\Gamma^\#$ ,  $f$  and  $\bar{f}$  for all relevant long lived chlorine and bromine species for a mean age of 5.5 years. In the case of fractional release factor of 1, there is no remaining organic fraction and the concept of mean arrival time  $\Gamma^*$  is not applicable (n.a.). Inorganic chlorine can then be treated in a similar way as an inert tracer, using mean age  $\Gamma$ . The time independent fractional release factors were derived using equation (32). The measurements from which the original fractional release factors were derived are from the period 1996-2000. We calculated the conversion using eq. (32) for every month of this period for all species, except for HCFC-141b and 142b (see text). The  $1\sigma$  variability of the converted fractional release factors is also shown.

	Stratospheric lifetime used for $\Gamma^*$ [years]	mean arrival time $\Gamma^*$ [years]	mean release time $\Gamma^\#$ [years]	fractional release factor $f$	time independent fractional release factor $\bar{f}$	$1\sigma$ variability of recalculated $\bar{f}$ in %
		Mean age $\Gamma$ of 5.5 years				
CFC-11	55.7 <sup>a</sup>	1.8	5.5	0.99 <sup>d</sup>	0.99	0.01 15
CFC-12	102 <sup>a</sup>	3.0	5.9	0.86 <sup>d</sup>	0.87	0.11
CFC-113	84.5 <sup>a</sup>	2.7	5.8	0.90 <sup>d</sup>	0.91	0.26
CFC-114	189 <sup>b</sup>	3.6	8.3	0.40 <sup>d</sup>	0.41	0.28 20
CFC-115	1020 <sup>b</sup>	4.4	10.1	0.15 <sup>d</sup>	0.20	0.58
CCl <sub>4</sub>	48.4 <sup>a</sup>	n.a.	5.5	1.00 <sup>d</sup>	1.00	0.00
CH <sub>3</sub> CCl <sub>3</sub>	48.8 <sup>a</sup>	1.7	5.6	0.99 <sup>d</sup>	0.99	0.13 25
HCFC-22	217 <sup>a</sup>	4.3	7.0	0.41 <sup>d</sup>	0.44	0.16
HCFC-141b	73 <sup>a</sup>	2.5	5.8	0.90 <sup>e</sup>	0.90	n.a. <sup>f</sup>
HCFC-142b	212 <sup>c</sup>	3.7	6.5	0.65 <sup>e</sup>	0.65	n.a. <sup>f</sup>
halon-1211	39.5 <sup>a</sup>	n.a.	5.5	1.00 <sup>d</sup>	1.00	0.00 30
halon-1202	15.3 <sup>c</sup>	n.a.	5.5	1.00 <sup>d</sup>	1.00	0.00
halon-1301	70.8 <sup>c</sup>	2.2	6.2	0.80 <sup>d</sup>	0.83	0.77
halon-2402	33.8 <sup>c</sup>	n.a.	5.5	1.00 <sup>d</sup>	1.00	0.00 35
CH <sub>3</sub> Br	40.2 <sup>a</sup>	1.5	5.5	0.99 <sup>d</sup>	0.99	0.01
CH <sub>3</sub> Cl	63.7 <sup>a</sup>	2.9	5.8	0.91 <sup>d</sup>	0.91	0.00 <sup>g</sup>

40 <sup>a</sup> stratospheric lifetime from the 2D model used ~~in~~ by Plumb et al. (1999)

<sup>b</sup> total atmospheric lifetime taken from recommendations in SPARC lifetime assessment (Ko et al., 2013), as tropospheric loss is negligible.

<sup>c</sup> stratospheric lifetime taken from modelling work for SPARC lifetime assessment (Chipperfield et al., 2013).

<sup>d</sup> fractional release values based on Newman et al. (2007)

45 <sup>e</sup> fractional release value based on parameterization given in footnote to table 2 in Velders and Daniel (2014).

<sup>f</sup> not applicable, as the fractional release values have not been recalculated.

<sup>g</sup> no variability was derived as there is no trend in the tropospheric reference data applied.

**Table 3:** Recovery years for EESC to return to 1980 values and maximum EESC values using our new formulation and the current formulation (Newman et al., 2007), as shown in Figures 3 and 4. In all cases the width  $\Delta$  is parametrized based a values of  $\lambda = \frac{\Delta^2}{\Gamma} = \frac{\Delta^{#2}}{\Gamma^{#}} =$  **0.7 years** (see text for an explanation of the parameterization). In the new formulation, the time-independent fractional release values and the values for  $\Gamma^{#}$  **shown in Tables A1 and A2** have been used. The results in the bottom row were derived using the same fractional release factors as in (Velders and Daniel, 2014), instead of the new time-independent fractional release factors, as shown in Figure 5 for 3 years of mean age. Decimal places are not meant to imply that recovery dates can be calculated to this accuracy rate, but are only given in order to show the sensitivity of the calculations to different parameters.

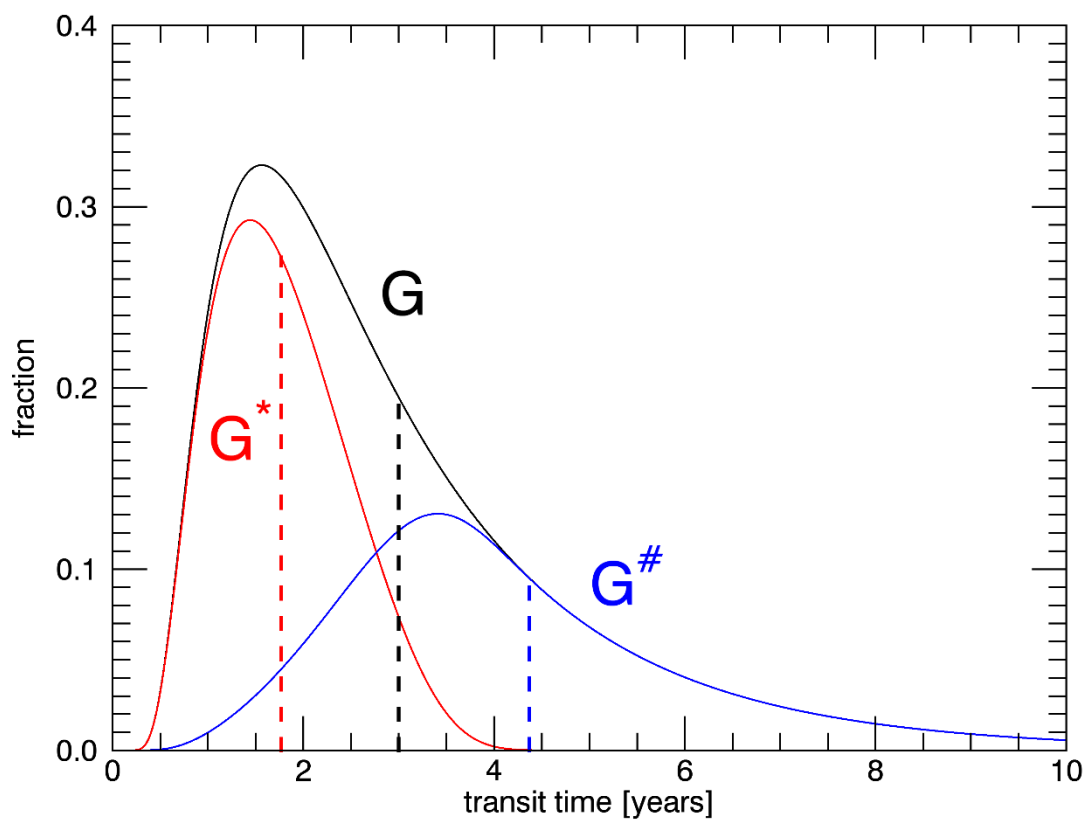
	3 years mean age			5.5 years mean age		
	EESC 1980 [ppt]	EESC max [ppt]	1980 recovery date	EESC 1980 [ppt]	EESC max [ppt]	1980 recovery date
New formulation, time independent fractional release factors (Table A1 and A2) and $G^{#}$ .	1065	1909	2059.9	2070	4107	2077.5
Current formulation, fractional release factors as in Velders and Daniel (2014), age spectrum G.	1154	1932	2048.6	2085	4102	2075.7
New formulation using $G^{#}$ , but unchanged fractional release factors as in Velders and Daniel (2014)	1070	1895	2057.8	2066	4088	2077.1

**Table 4:** Recovery years for EESC to return to 1980 values and maximum EESC values using different assumptions on the width of the release time distribution  $G^{#}$  as shown in Figure 6 and 7. In all cases the general shape was assumed to be an inverse Gaussian function with different parameterizations of the width  $\Delta$ , based on different values of  $\lambda = \frac{\Delta^{#2}}{\Gamma^{#}}$  (see text for an explanation of the parameterization). The case of the pure lag time calculation is equal to  $\lambda = 0$  years, i.e. the effect of mixing is completely ignored,  $\lambda = 2$  years represents a case with strong mixing and a broad transit time distribution, while,  $\lambda = 0.7$  years corresponds to our reference calculation. In all calculations, the same time-independent fractional release values have been used, as shown in Tables A1 and A2. Decimal places are not meant to imply that recovery dates can be calculated to this accuracy rate, but are only given in order to show the sensitivity of the calculations to different parameters.

	3 years mean age			5.5 years mean age		
	EESC 1980 [ppt]	EESC max [ppt]	1980 recovery date	EESC 1980 [ppt]	EESC max [ppt]	1980 recovery date
$\lambda = 0.7$ years reference	1065	1909	2059.9	2070	4107	2077.5
$\lambda = 2$ years, strong mixing	1101	1913	2058.5	2149	4103	2075.0
$\lambda = 0$ , no mixing	1055	1941	2060.7	2046	4176	2078.8

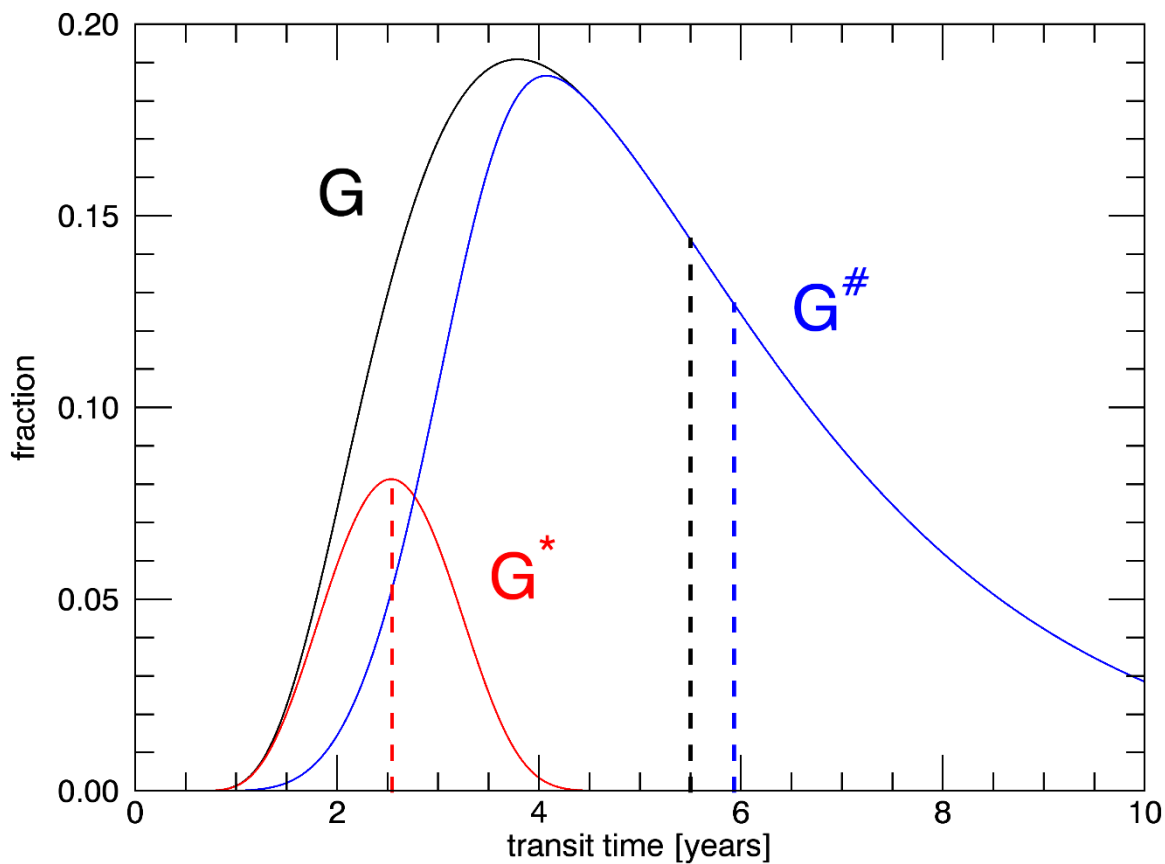
**Table 5:** Recovery years for EESC to return to 1980 values and maximum EESC values varying the stratospheric lifetimes in the calculations of the mean arrival time  $\Gamma^*$  from which the mean release time  $\Gamma^\#$  is calculated according to (A8). In these calculations, the width of the release time distribution  $G^\#$  was kept constant at  $\lambda = \frac{\Delta^\#{}^2}{\Gamma^\#} = 0.7$  years. The lifetimes have been varied systematically up- and downward by 20% for this sensitivity test (see text for explanations). The reference case is the same as shown in Table 1. For the high  $\tau$  and low  $\tau$  calculations the stratospheric lifetimes of all species have been increased systematically by 20% upwards and downwards respectively, when calculating the mean arrival times and the mean release times. Decimal places are not meant to imply that recovery dates can be calculated to this accuracy rate, but are only given in order to show the sensitivity of the calculations to different parameters.

	3 years mean age			5.5 years mean age		
	EESC 1980 [ppt]	EESC max [ppt]	1980 recovery date	EESC 1980 [ppt]	EESC max [ppt]	1980 recovery date
Reference	1064	1909	2059.9	2070	4107	2077.5
High $\tau$	1073	1912	2058.8	2071	4107	2077.3
low $\tau$	1054	1906	2061.3	2068	4107	2077.6

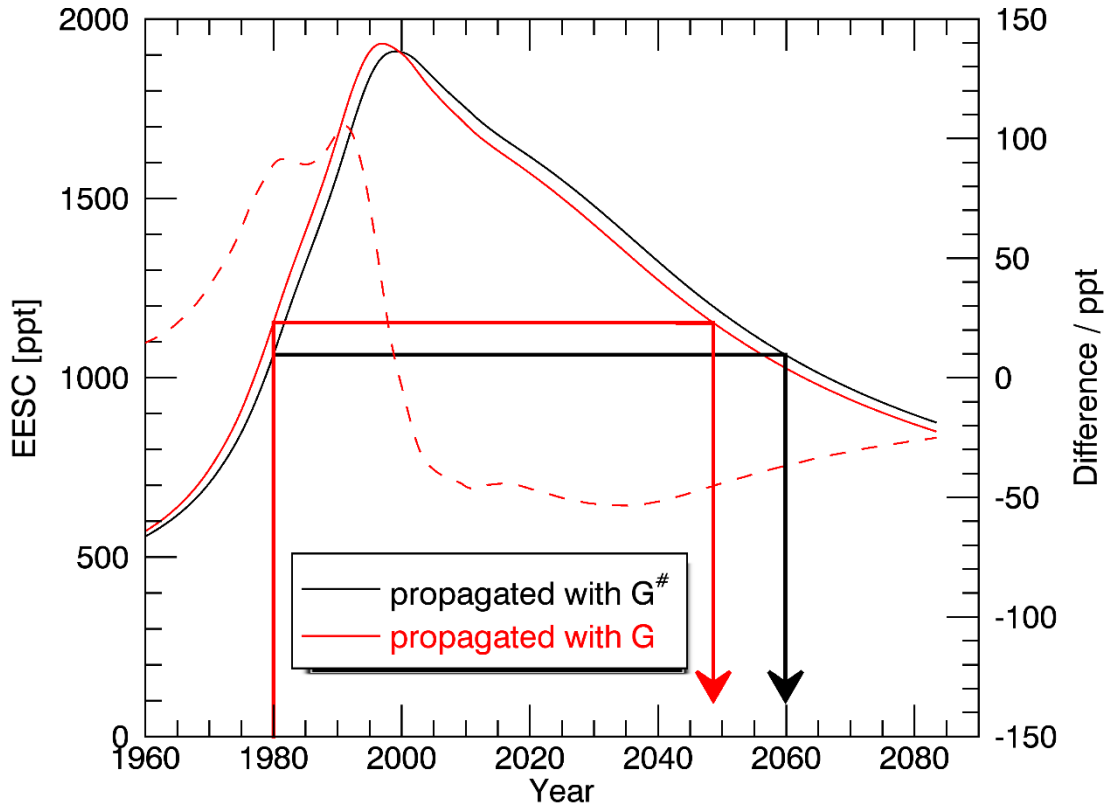


5 **Figure 1:** Age spectrum  $G$  (black line) for an inert tracer compared to the arrival time distribution  $G^*$  (red line) and the release time distribution  $G^\#$  (blue line). The loss function has been approximated as a function of transit time in order to represent a tracer similar to CFC-11 (see supplementary information for more details). The first moments of the three functions differ substantially: while the black line (inert tracer) has a first moment of 3 years, the first moment for the red curve, representing the remaining organic fraction is 1.75 years and that of the blue curve describing the inorganic halogen released from the source gas is 4.35 years. Note that these values are not identical to those for CFC-11 in Table A1 as the loss function was only approximated and that this Figure is purely for illustrative purposes.



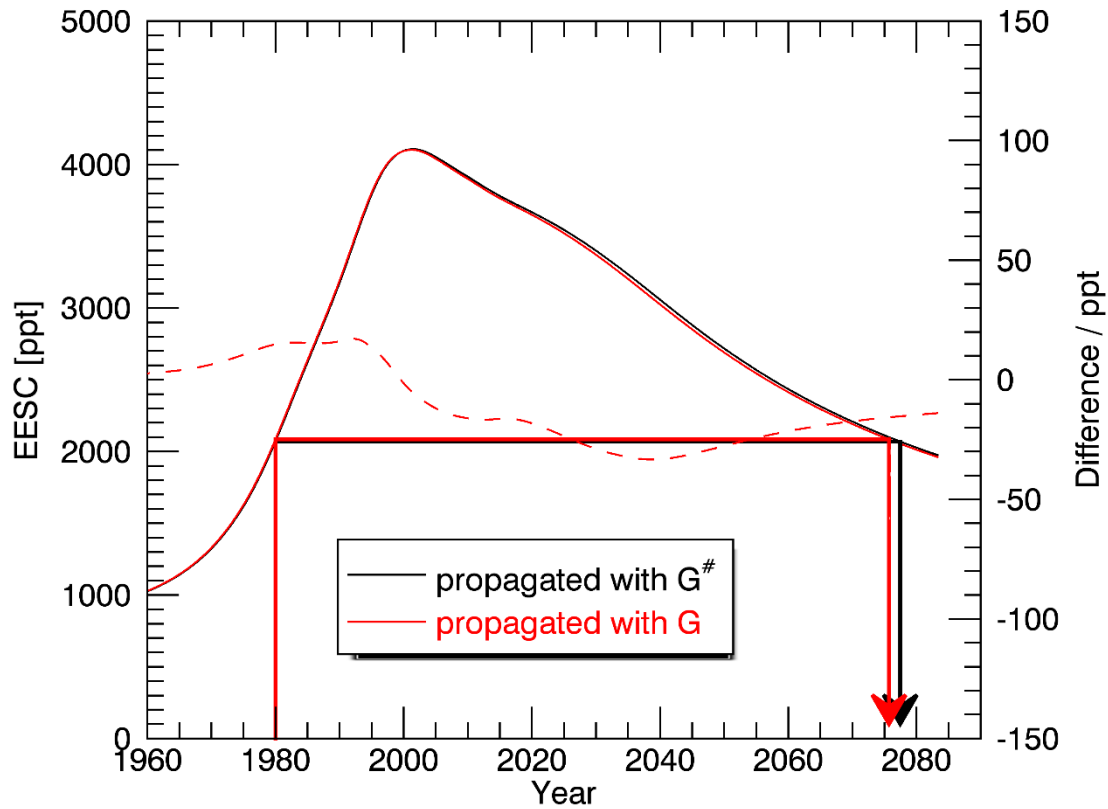


5 **Figure 2:** Age spectrum  $G$  (black line) for an inert tracer compared to the arrival time distribution  $G^*$  (red line) and the release time distribution  $G^\#$  (blue line). The loss function has been approximated as a function of transit time in order to represent a tracer similar to CFC-11 (see supplementary information for more details). The first moments of the three functions differ substantially: while the black line (inert tracer) has a first moment of 5.5 years, the first moment for the red curve, representing the remaining organic fraction is 2.5 years and that of the blue curve describing the inorganic halogen released from the source gas is 5.9 years. Note that these values are not identical to those for CFC-11 in Table A2 as the loss function was only approximated and that this Figure is purely for illustrative purposes.

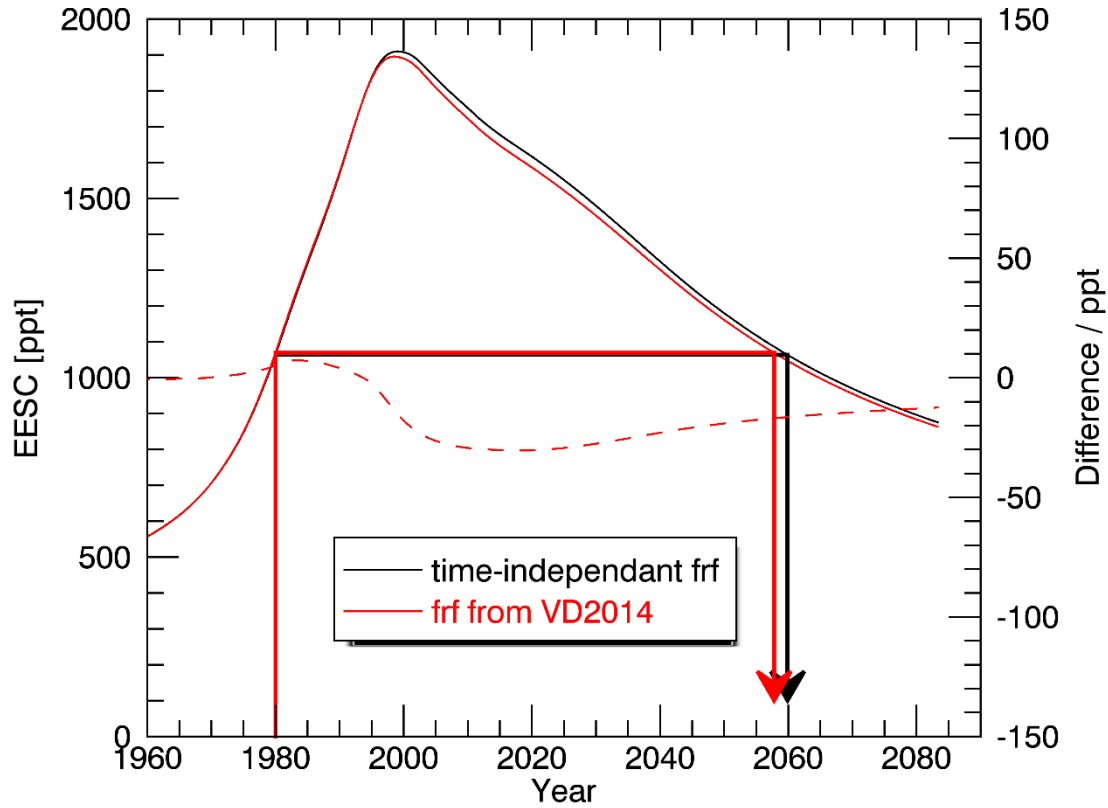


**Figure 3:** Estimated temporal evolution of EESC for a mean age of 3 years using the old (red line) and the new (black line) formulation of EESC. Also shown is the difference (red dashed line) and the recovery date to 1980 values for the old and the new formulation. Our new formulation yields a recovery date, which is more than 10 years later than using the current formulation. This shift in recovery date is mainly caused by the lower EESC levels calculated for the increasing phase, i.e. the 1980 reference value.

5



**Figure 4:** Estimated temporal evolution of EESC for a mean age of 5.5 years using the old (red line) and the new (black line) formulation of EESC. Also shown is the difference (red dashed line) and the recovery date to 1980 values for the old and the new formulation. Our new formulation yields a recovery date to 1980 values, which is about 2 years later than using the current formulation. The smaller shift in comparison to the calculation for 3 years of mean age is due to the near complete fractional release of most halogen source gases for these old air masses.



**Figure 5:** Influence of new fractional release factors for calculation of EESC at a mean age of 3 years. In both calculations the new formulation of EESC has been used, yet both calculations use different fractional release factors. The calculation using the new fractional release factors (Table A1) for 3 years of mean age is shown in black, while the calculation using the original values as used ~~in~~by Velders and Daniel (2014) (VD2014) is shown in red. As the fractional release factors currently used (Harris et al., 2014; Velders and Daniel, 2014) are largely based on measurements (Newman et al., 2007; Schauffler et al., 2003), which were taken during a period of rather small tropospheric trends for most species, the change due to the new formulation (Ostermüller et al., 2017) is rather small. For HCFCs 141b and 142b, uncertainties on observational fractional release factors are large and the same fractional release factors were used in both calculations, which are based on the parameterization given ~~in~~by Velders and Daniel (2014). The difference of the calculation using the VD2014 fractional release factors and our new time independent fractional release factors is shown as red dashed line. The fractional release factors used here are summarized in Table A1.

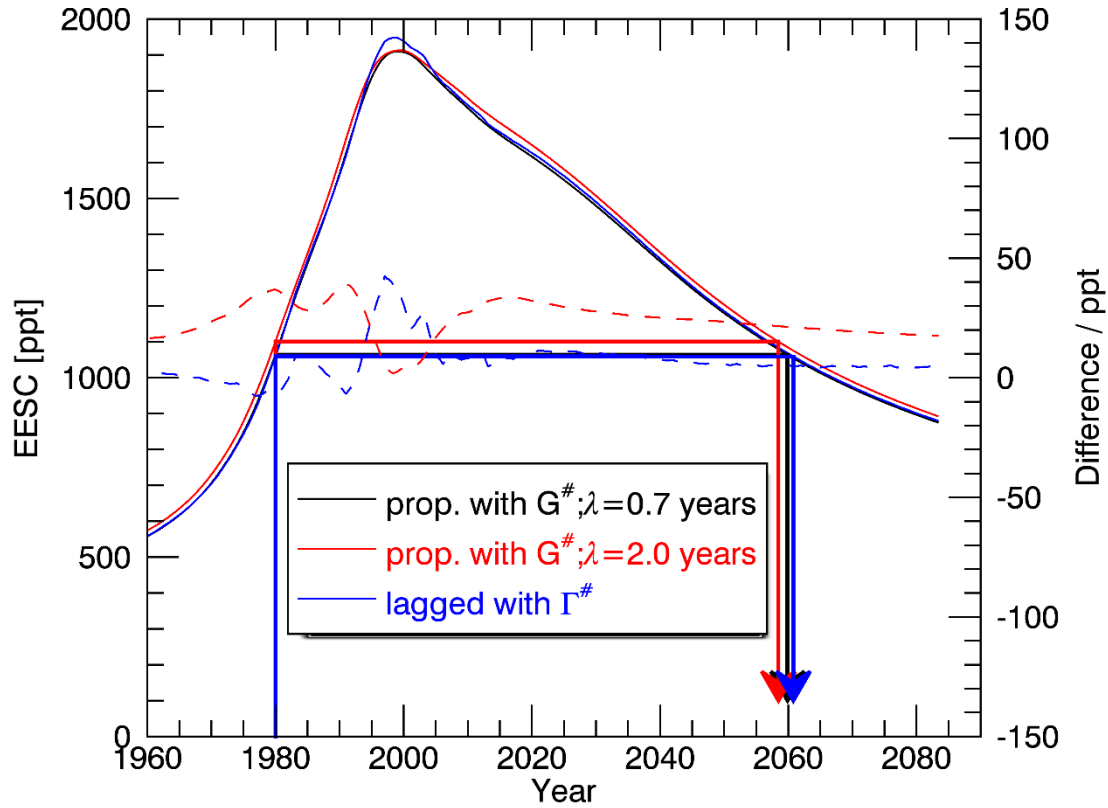
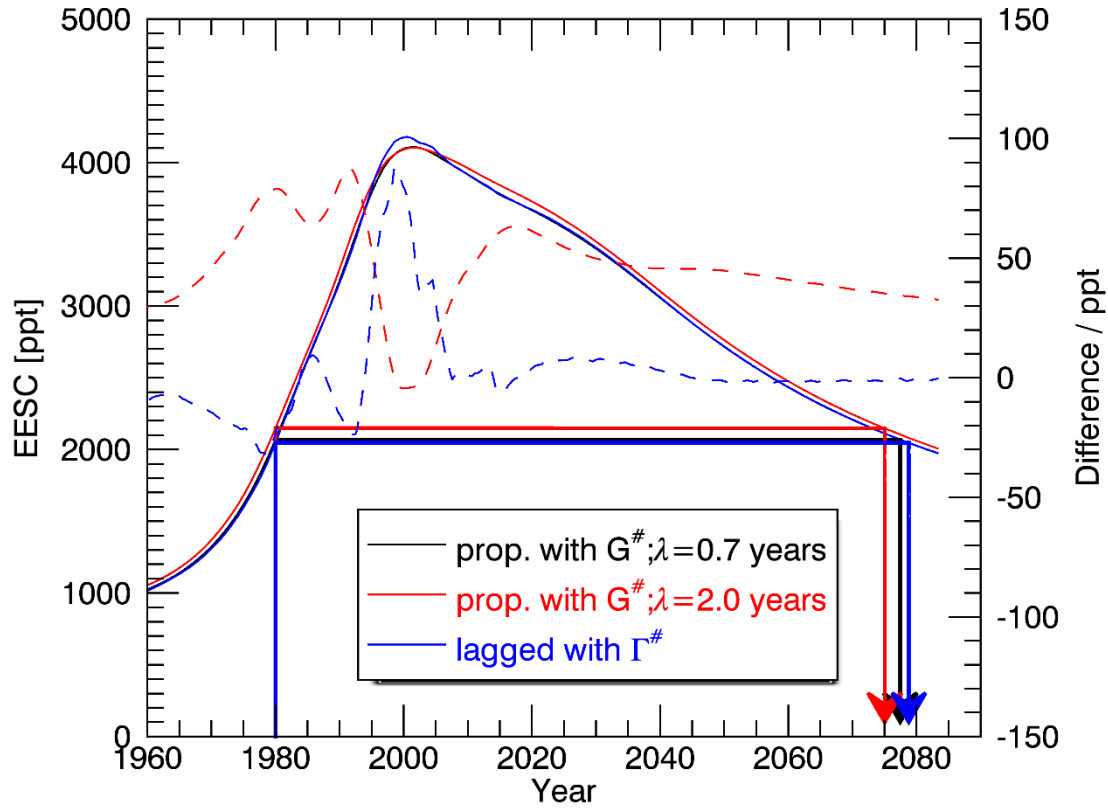
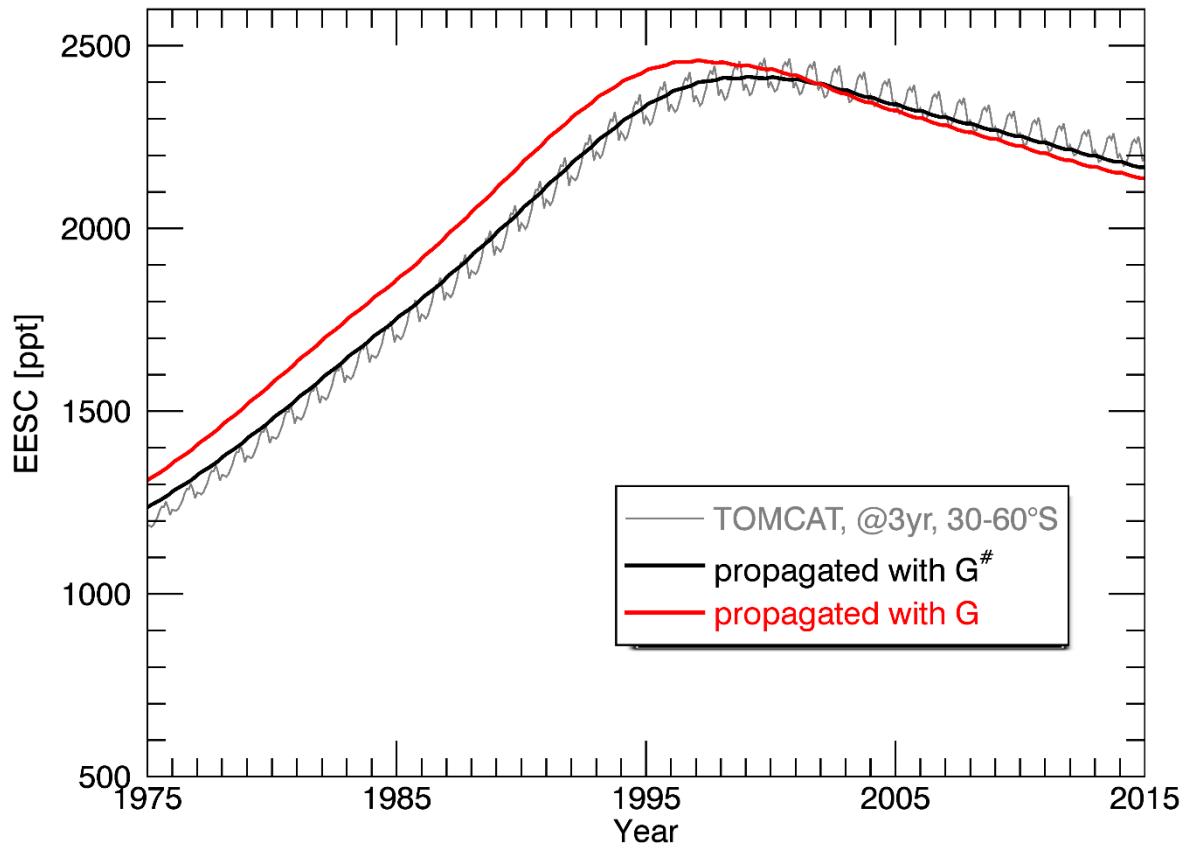


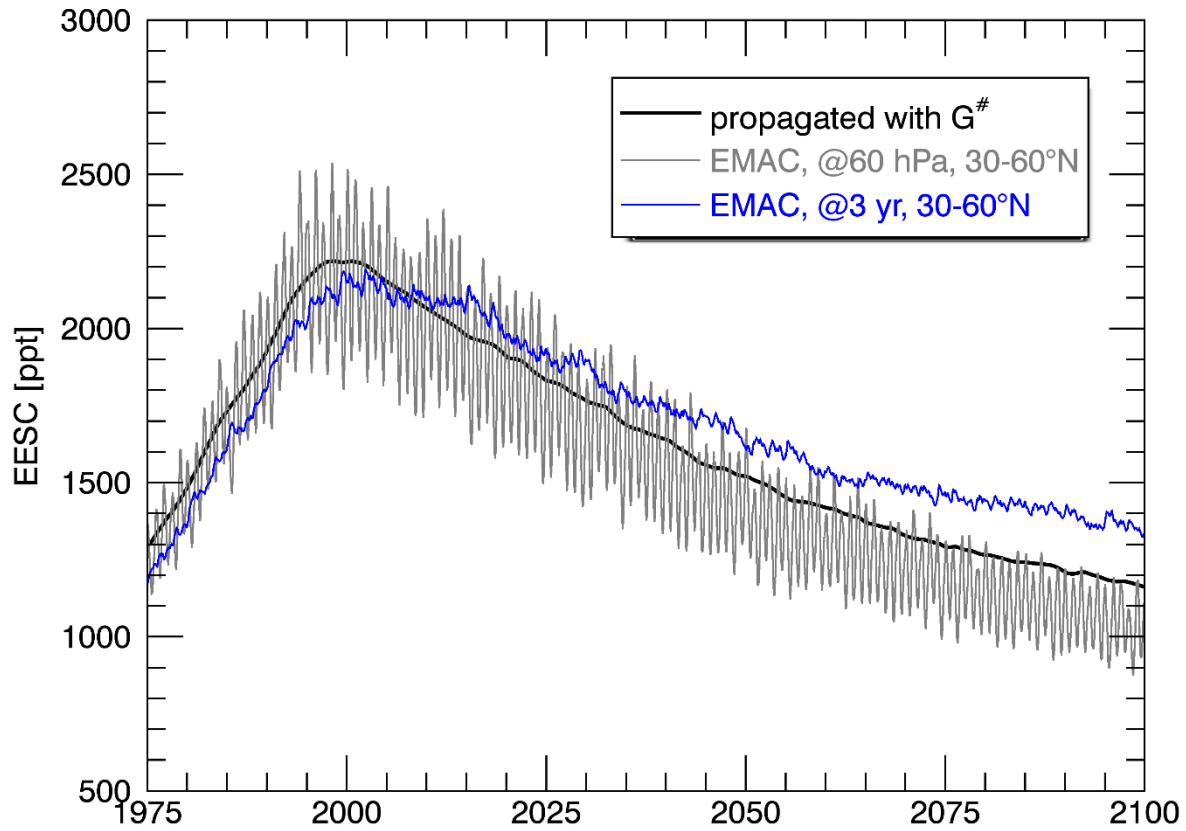
Figure 6: Sensitivity of EESC calculation using the new formulation for a mean age of 3 years on the parameterization of the shape of the release time distribution. In two cases the general shape was assumed to be an inverse Gaussian function with different parameterizations of the width  $\Delta$ , based on different values of  $\lambda = \frac{\Delta^2}{\Gamma}$  (see text for an explanation of the parameterization). The case of the pure lag time calculation (blue line) is equal to  $\lambda = 0$  years, i.e. the effect of mixing is completely ignored. The calculation using  $\lambda = 2$  years (red line) represents a case with strong mixing and a broad transit time distribution, while  $\lambda = 0.7$  years (black line) corresponds to our reference calculation. The influence is largest during the period of the maximum, when tropospheric trends showed a strong non-linear behavior and the tracer propagation strongly depends on the shape of the distribution function. The difference of the calculation to our reference calculations are shown as dashed lines.



**Figure 7:** Sensitivity of EESC calculation using the new formulation for a mean age of 5.5 years on the parameterization of the shape of the release time distribution. In two cases the general shape was assumed to be an inverse Gaussian function with different parameterizations of the width  $\Delta$ , based on different values of  $\lambda = \frac{\Delta^2}{\Gamma}$  (see text for an explanation of the parameterization). The case of the pure lag time calculation (blue line) is equal to  $\lambda = 0$  years, i.e. the effect of mixing is completely ignored. The calculation using  $\lambda = 2$  years (red line) represents a case with strong mixing and a broad transit time distribution, while  $\lambda = 0.7$  years (black line) corresponds to our reference calculation. The influence is largest during the period of the maximum, when tropospheric trends showed a strong non-linear behavior and the tracer propagation strongly depends on the shape of the distribution function. The difference of the calculations to our reference calculation are shown as dashed lines.



**Figure 8:** Comparison of EESC using the formulation by (Newman et al., 2007) and the new formulation suggested here to TOMCAT model calculations (Chipperfield et al., 2017) of ESC for Southern Hemisphere mid latitude conditions (3 years mean age). Fractional release values were calculated from the model and differ from those shown in Table 1, but are used in order for EESC and ESC to be consistent. The model simulation used here has fixed dynamics, using 1980 meteorology. While small differences remain, the new formulation yields much better agreement between EESC and ESC.



**Figure 9:** Comparison of EESC at 3 years of mean age using our new formulation to model ESC evaluated at the 60 hPa level (corresponding to 3 years of mean age in 2000) and to ESC at 3 years of mean age. Model data are from the EMAC model as described by Jöckel et al. (2016). Fractional release values were calculated from the model and differ from those shown in Table 1, but are used in order for EESC and ESC to be consistent. The model simulation used here uses prescribed trace gas scenarios and prescribed sea surface temperatures and sea ice content.