



Mixing ratios, EESC,
ODPs, and GWPs

G. J. M. Velders and
J. S. Daniel

Uncertainty analysis of projections of ozone-depleting substances: mixing ratios, EESC, ODPs, and GWPs

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Abstract

The rates at which ozone depleting substances (ODSs) are removed from the atmosphere, that is, their lifetimes, are key factors for determining the rate of ozone layer recovery in the coming decades. We present here a comprehensive uncertainty analysis of future mixing ratios of ODSs, levels of equivalent effective stratospheric chlorine (EESC), ozone depletion potentials, and global warming potentials, using, among other information, the 2013 WCRP/SPARC assessment of lifetimes of ODSs and their uncertainties. The year EESC returns to pre-1980 levels, a metric commonly used to indicate a level of recovery from ODS-induced ozone depletion, is 2048 for mid-latitudes based on the lifetimes from the SPARC assessment, which is about 2 yr later than based on the lifetimes from the WMO assessment of 2011. However, the uncertainty in this return to 1980 levels is much larger than the 2 yr change. The year EESC returns to pre-1980 levels ranges from 2039 to 2064 (95 % confidence interval) for mid-latitudes and 2061 to 2105 for the Antarctic spring. The primary contribution to these ranges comes from the uncertainty in the lifetimes. The earlier years of the return estimates are comparable to a hypothetical scenario in which emissions of ODSs cease in 2014. The later end of the range corresponds to a scenario containing an additional emission of about 7 MtCFC-11-eq in 2015, which is the same as about 2 times the cumulative anthropogenic emissions of all ODSs from 2014 to 2050, or about 12 times the cumulative HCFC emissions from 2014 to 2050.

1 Introduction

Projections of the mixing ratios of ozone-depleting substances (ODSs) and the levels of chlorine and bromine in the stratosphere require knowledge of future emissions and loss processes in addition to current atmospheric abundances and amounts of ODSs present in existing equipment. In the past five WMO/UNEP Scientific Assessments of Ozone Depletion (WMO, 1995, 1999, 2003, 2007, 2011) a box model has been

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used to calculate mixing ratios and equivalent effective stratospheric chlorine (EESC) from historic data and future projections of ODSs. EESC is an index that relates the time evolution of long-lived ODSs in the troposphere with their ability to destroy stratospheric ozone (Daniel et al., 1995, 2010; WMO, 2011). Uncertainties in parameters and their effects on mixing ratios and EESC levels have not received much attention Newman et al. (2007) reformulated EESC by using the age-of-air spectrum and age-of-air dependent fractional release values and analyzed the effects of uncertainty in these parameters on EESC. They found that the largest uncertainties in EESC are associated with the mean age-of-air in the stratosphere and the fractional release of halogens from ODSs in the stratosphere. Daniel et al. (2007) studied the effects of different assumptions for the amounts of ODSs present in existing equipment on future EESC levels. One of the key pieces of information for calculating future mixing ratios and EESC and for estimate past ODS emissions is knowledge of the ODS lifetimes; yet, to our knowledge, an uncertainty analysis has not been performed with respect to lifetime uncertainties. Douglass et al. (2008) suggested that past estimates of the CFC-11 lifetime were too short, and that part of the cause was that only recently have models begun to have a slow enough stratospheric circulation to be consistent with age-of-air observations. The WMO/UNEP assessment (WMO, 2011) indeed concluded that there is emerging evidence that the lifetimes of some important ODSs (e.g. CFC-11) may be somewhat longer than previously reported (Montzka et al., 2011b). Since then several papers have been published on lifetimes of ODSs (Laube et al., 2013; Minschwaner et al., 2013; Rigby et al., 2013; Brown et al., 2013) and an assessment of lifetimes has been conducted by the World Climate Research Programme (WCRP) Stratospheric Processes And their Role in Climate (SPARC) project (SPARC, 2013). In the SPARC assessment, a new set of recommended lifetimes are presented based on calculations with atmospheric chemistry-transport models, analysis of observations at the surface and in the stratosphere, laboratory analysis of chemical reactions and photolysis rates, and on inverse modelling. Apart from a new set of lifetimes, a complete uncertainty analysis of lifetimes of all the major halogenated ODSs is presented for the

first time. The uncertainties in the lifetimes are considerable, ranging from 3 % to 33 % (1 σ), with significant implications for projecting the decay of ODS concentrations in the atmosphere. The new lifetimes and associated uncertainties and their effects on future EESC levels are the basis of the analysis presented here.

Four items are explored. First, the effects of the new SPARC (2013) lifetimes on future mixing ratios of ODSs and EESC levels are compared with the values calculated with the lifetimes from WMO (2011). Second, the effects of the uncertainties in the lifetimes on mixing ratios and EESC levels are discussed. Third, a complete uncertainty analysis is performed for the calculations of EESC levels and the year of return to 1980 EESC levels as well as for estimates of ODS radiative forcing, by considering uncertainties in all the relevant parameters. Fourth, Ozone Depletion Potentials (ODPs), Global Warming Potentials (GWPs), and their uncertainties are calculated using the new lifetime information.

Here we focus on the long-lived chlorine and bromine containing species and the EESC as a measure of the amount of active chlorine/bromine in the stratosphere available to cause ozone depletion. We recognize that other factors are also important for the future development of the ozone layer, such as future concentrations of nitrous oxide (N₂O), methane (CH₄), very short-lived halogen containing species, and changes in temperature and circulation in the atmosphere. These other factors are not considered here.

The model, input data, lifetimes, and uncertainties in input parameters are discussed in Sect. 2. The results of the box model calculations, the effects of the uncertainties in the lifetimes and other parameters on mixing ratios and EESC are presented in Sect. 3, followed by uncertainties in radiative forcing of ODSs in Sect. 4. In Sect. 5 and 6 uncertainties in ODPs and GWPs are presented. A discussion of the results and conclusions follows in Sect. 7.

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2 Model and input description

2.1 Box model

For the calculation of mixing ratios and EESC almost the same box model and data are used as in the Scientific Assessments of Ozone Depletion: 2010 (WMO, 2011). There are only a few minor exceptions. One difference is the use of the age-of-air spectrum from Newman et al. (2007) and age-of-air dependent fractional release values from Newman et al. (2006) in the calculation of EESC. These fractional release values have also been used in WMO (2011), but only for discrete ages-of-air for mid-latitude (3 yr) and Antarctic (5.5 yr) conditions. These changes in methodology are necessary for the Monte Carlo uncertainty analysis. There are also a few minor differences even when using ages of 3 and 5.5 yr due to rounding that was performed in WMO (2011).

In the box model, average surface mixing ratios of ODSs are calculated from mean atmospheric mixing ratios using a fixed factor. The EESC is calculated for mid-latitude and Antarctic conditions from these mixing ratios. The surface global mean mixing ratio of specific ODS i is calculated using Eq. (1),

$$\frac{d\rho_i}{dt} = F_i E_i - \frac{\rho_i}{\tau_i} \quad (1)$$

where ρ_i is the global mean mixing ratio (in ppt), τ_i is the total lifetime (in yr), E_i is the emission rate (in kg yr^{-1}). F_i (in ppt kg^{-1}) is a factor that relates the mass emitted to the global mean mixing ratio as expressed by Eq. (2),

$$F_i = \frac{N_A F_{\text{surf}}}{M_i N_{\text{atm}}} = \frac{F_{\text{surf}} \cdot 5.68 \times 10^{-9}}{M_i} \quad (2)$$

where M_i is the molecular weight (in kg mol^{-1}), N_A the Avogadro constant, N_{atm} the number of molecules in the global atmosphere, and F_{surf} is a factor relating the global mean atmospheric mixing ratio to the global surface mixing ratio. The factor F_{surf} is

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meant to account for the general decrease of the ODS mixing ratios with altitude above the tropopause. In WMO (2011), F_{surf} was taken to be 1.07 for all ODSs, except for CH_3Br for which 1.16 was taken. Using a constant for F_{surf} neglects change in this factor that could be caused by changes in circulation or by the variability of the surface emission (and the resulting variability in the atmospheric vertical distribution).

For the emissions in the model a different approach is used for historic emissions than for future emissions. As in WMO (2011), historical annual emissions are derived, using Eq. (1), from observed mixing ratios and the lifetimes of the ODSs. This so called top-down approach, is taken, because the ODSs are long-lived and the global mixing ratios and lifetimes are thought to be known more accurately than are independently derived emissions. Future emissions are estimated from a scenario of future production and the amounts of ODSs present in existing equipment and applications, generally called “banks”. Banks here are defined as the quantity of ODSs produced but not yet emitted to the atmosphere. These quantities will be present mostly in existing refrigeration and air conditioning units and foams, but can also be in landfills. The bank sizes for a specific year (2008 here) are based on so called bottom-up estimates from inventories of equipment that contains a particular ODS (IPCC/TEAP, 2005; TEAP, 2009). With annual reported production and top-down derived emissions the bank can be calculated backwards in time starting with the bottom-up bank of 2008. For years further before 2008, it is expected that bank uncertainties will increase because the bank difference is a cumulative difference of two large numbers, namely production and emission. However, for our analysis purposes, this uncertainty only affects our results through the bank emission factor. The annual emissions after 2008 are assumed to be a constant fraction of the bank. This bank emission factor (E_f) is determined as the fraction of the top-down derived emissions and the banks averaged over the previous ten years.

The following step-by-step approach is used to obtain a mixing ratio time series from 1950 to 2120. First, top-down emissions are derived from observed mixing ratios and lifetimes. Second, bank sizes are calculated backwards starting in 2008 using reported production data and top-down emissions. Third, emission factors for future emissions

are calculated as the 10 yr average of the fraction of the emission over the bank (1999–2008). Fourth, projections of mixing ratios are calculated using the box model (Eq. 1) out to 2120. Emissions and mixing ratios before 1979 were derived from bottom-up emission estimates (WMO, 2003, 2007, 2011).

EESC is calculated using these mixing ratios, fractional release values, which account for the differences among ODSs in their efficiency to release halogens in the stratosphere, a time lag (age-of-air) for air to be transported from the surface to the stratosphere, and a factor α to account for the relative effectiveness of bromine compared with chlorine for ozone depletion. A value of 60 is used for α for mid-latitude conditions and 65 for Antarctic conditions (WMO, 2007, 2011). The single values for the 3 yr age-of-air for mid-latitude conditions and 5.5 yr for Antarctic conditions, used in WMO (2011), are replaced by age-of-air spectra from Newman et al. (2007) with the same mean ages as before. The fractional release values of the ODSs corresponding to 3 and 5.5 yr mean age-of-air are replaced by functions describing the age-of-air dependence of the fractional releases (Newman et al., 2006). These functions yield the same values for 3 and 5.5 yr age-of-air as those used in WMO (2011), except for two. As argued in WMO (2011), the abundances of HCFC-141b and HCFC-142b were small and had large temporal trends at the time the measurements upon which these fractional release values are based were made, leading to large uncertainties. Therefore, the functions describing the age-of-air dependent fractional release values for HCFC-141b and HCFC-142b were not used in WMO (2011), but instead values from WMO (2007) were used. Here, age-of-air dependent fractional release values for HCFC-141b and HCFC-142b are used by adjusting the functions of Newman et al. (2006) to be in better agreement with the values used in WMO (2011, 2007) for 3 and 5.5 yr mean ages and with the curves of other ODS (see footnote Table 2).

Recently new fractional release values with associated uncertainties have been reported by Laube et al. (2013). The absolute values derived by Laube et al. (2013) are about 20 % smaller than those derived by Newman et al. (2006), and those used in WMO (2011). Although this 20 % does result in smaller EESC levels, it hardly changes

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the EESC curve shape, and the year EESC returns to pre-1980 levels is only changed by about one year for mid-latitude conditions. For Antarctic spring conditions the year EESC returns to pre-1980 levels is about five years earlier with the fractional release values from Laube et al. (2013) compared with those from WMO (2011). As discussed by Laube et al. (2013), the differences in fractional release values are probably caused by differences in the determination of the age-of-air in the measurements and by inter-annual variability in the fractional release of the halogens. Since both sets of fractional release values yield about the same EESC time series, we choose to use the recommended fractional release values from WMO (2011). Thus, in our uncertainty analysis for EESC, only the Laube et al. (2013) fractional release uncertainties have been used, not the absolute fractional release values. The fractional release values from Laube et al. (2013) have been applied for sensitivity analyses in the calculation of ODPs (Sect. 5).

Following the past ozone assessments, two quantities are derived from the EESC time series, namely, the year EESC returns to a pre-1980 level and the integrated EESC above the 1980-level. These quantities have been used extensively to compare different ODS scenarios.

2.2 ODS production, banks, and observed mixing ratios

We use production data from 1950 to 2008 that have been officially reported by the countries to UNEP. For the production from 2009 and after, the data from the baseline scenario of WMO (2011) is used. There is no known production anymore of CCl₄ for its historical primary uses (e.g., as a solvent) and also no corresponding bank, but there are still considerable emissions as derived from observed mixing ratios. Therefore, following WMO (2011), relative annual change in emission from 2004–2008 is extrapolated for 2009 to 2050. As a consequence of this approach, these future emissions depend on the lifetime of CCl₄ since it is a key factor for deriving its top-down emissions over the last few years. This uncertainty in the source of current CCl₄ emissions also suggests that future CCl₄ emissions are more uncertain than most of the other ODSs;

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because it is not clear how to quantify this extrapolation uncertainty, it is not included in our uncertainty analysis.

In addition to the baseline scenario, a zero emissions (from 2014 onward) scenario (see also WMO, 2011) is used to put the results in perspective of the maximum mitigation that could be done for ODSs, barring active removal from the atmosphere.

As described in Sect. 2.1 the bank of ODSs for the year 2008 is used as a constraint for the calculations with the model. These bottom-up estimates are derived from inventories of the number of units of equipment that use a particular ODS and the amount of ODS in each unit. The bank sizes used here are those used in WMO (2011) and reported by TEAP (2009).

Another constraint for the model calculations is the observed mixing ratios, which are used to derive top-down emission estimates and as a starting point for the mixing ratio projections. The observations up to the beginning of 2009 are from WMO (2011) for all ODSs. For most species, they are based on the average of the annual global mean values from the AGAGE and NOAA/ESRL networks.

2.3 Lifetimes of ODSs

The lifetimes of ODSs are estimated from their loss rates and play an important role in the model calculations. A number of the ODSs, all CFCs, CCl_4 and Halon-1301, are removed primarily by photolysis in the stratosphere (SPARC, 2013). The lifetimes of these ODSs depend on the absorption cross sections of the ODSs, solar irradiances, and oxygen and ozone opacities. The dependence on the solar irradiances and opacities means that the uncertainties in these ODS photolysis rates, and consequently the lifetimes, are strongly correlated.

Other ODSs, all HCFCs, CH_3CCl_3 , Halon-1211, -1202 and -2402, CH_3Br and CH_3Cl , are removed primarily in the troposphere by reaction with the hydroxyl radical (OH), although a significant fraction of Halon-1211 and Halon-2402 is removed in the stratosphere (SPARC, 2013). Therefore, the uncertainty in the lifetime of these ODSs de-

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depends on the uncertainty in the OH radical abundance in the troposphere. The uncertainties of these ODSs is therefore also strongly correlated.

In the model calculations the total steady-state lifetimes as reported by WMO (2011) are used as well as those reported by SPARC (2013). The effect of the differences in lifetimes on future mixing ratios and EESC is examined (Sect. 3.1).

Simulations with several 2-D and 3-D atmospheric chemistry-transport models reported by SPARC (2013) show instantaneous lifetimes from 1960 to 2010 that deviate somewhat from the steady-state lifetimes. Instantaneous lifetimes of most ODSs decrease from 1960 to 2010, most dramatically in the early years of their use, mainly as a result of trends in atmospheric concentrations from an imbalance between emissions and loss rates (Martinerie et al., 2009). The model calculations show only a small trend in lifetimes from 2010 to 2100. These time-varying instantaneous lifetimes have not been used in the calculations here, but their effects on mixing ratios or inverse derived emissions is small. The instantaneous lifetimes of CFC-11 and CFC-12 are about 50 % larger than the steady-state lifetimes around 1960, decreasing to about 20 % larger around 1980 and smaller towards 2010. Annual emissions of CFC-11 and CFC-12 derived from observed mixing ratios using these instantaneous lifetimes are only 0 to 4 % smaller over the period 1960–2010 than those derived using the steady-state lifetimes.

2.4 Parameters for Monte Carlo uncertainty analysis

In performing the Monte Carlo analysis to estimate the total uncertainty in EESC and the estimated time to return to pre-1980 EESC levels, uncertainties are applied to all relevant parameters, including: (1) annual historic and future production, (2) bank size of 2008, (3) observed mixing ratios, (4) fraction of the bank emitted annually (E_f), (5) loss rate (inverse of the lifetime) of the ODSs, (6) fractional release values, (7) factor (α) for bromine efficiency in ozone depletion, (8) mean age-of-air, and (9) factor relating the average atmospheric mixing ratio to the value at the surface (F_{surf}). In calculating the radiative forcing and GWPs of ODSs an additional uncertainty in the radiative efficiency is taken into account. For α and the mean age-of-air, different parameters are

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used for mid-latitude and Antarctic conditions. See Tables 1 and 2 for an overview of all the uncertainties used. In SPARC (2013) two uncertainty ranges are given for the ODS loss rate and lifetime, a “possible range” and a “most likely” range. These ranges differ for ODSs for which different methods have been used to derive the loss rate, such as modeling with atmospheric chemistry-transport models, inverse modeling using observed mixing ratios, analysis of satellite data, and tracer-tracer studies. The possible range is derived from the joint distribution of the different methods and might be an overestimation of the true uncertainty, since it gives the range in the lifetime estimates. The most likely range is derived from the combined distribution of the different methods and represents the variability of the best estimate. This range might be an underestimation of the true uncertainty since, for example, it assumes the different methods are uncorrelated and that they are estimating the same quantity. Both uncertainty ranges are presented and discussed in our uncertainty analysis because the uncertainty in the lifetimes is the most important parameter in the analysis.

As mentioned in Sect. 2.3 the loss rates of the ODSs that are removed mainly in the stratosphere all depend on the oxygen and ozone opacity and solar irradiances. As a consequence, the uncertainty in the loss rates of these species is correlated. This correlation is taken into account in the Monte Carlo analysis by applying two random numbers to determine the loss rate of an ODS. A random variation of the loss rate of ODS i , denoted by $\sigma_r(L_i)$ is calculated using Eq. (3),

$$\sigma_r(L_i) = \sigma(L_i) \cdot \left(\text{random}(1) \cdot \gamma + \text{random}(2) \cdot \sqrt{1 - \gamma^2} \right) \quad (3)$$

where $\sigma(L_i)$ is the uncertainty in the loss rate and “random()” indicates a random number drawn from a normal distribution ($N(0,1)$). Correlation between loss rates $\sigma_r(L_i)$ of different ODSs is obtained by applying the same random number (random(1)) to all ODSs that are destroyed mainly by photolysis in the stratosphere. A default correlation coefficient γ of 0.9 is assumed here. Such a coefficient means that 67 % of the random variation of the uncertainty in the loss rate of an ODSs in Eq. (3) is associated with

common uncertainties (OH mixing ratio for species removed mainly in the troposphere and solar irradiance and opacity for species mainly removed in the stratosphere) and 33 % with the specific ODSs. The sensitivity of EESC to the value of γ is studies in Sect. 3.2.

The same approach is used for the ODSs that are mainly removed in the troposphere by reaction with the OH radical. The uncertainties in the loss rates of these species is also correlated using Eq. (3) with the same correlation coefficient, but with different random numbers.

The fractional release values depend on the age-of-air following Newman et al. (2006). In addition to the uncertainty in the mean age-of-air an additional uncertainty of ± 10 to ± 20 % (1σ) is applied to the fractional release values of all ODSs (Table 2, Laube et al., 2013).

Uncertainty in global production data is assumed to be small. The historic production data is reported by a small number of companies and officially reported by countries to UNEP (UNEP, 2008). An uncertainty of ± 5 % (1σ) is assumed in the annual historic and future production data. It is assumed that the data is 100 % correlated between successive years, but completely uncorrelated among species. In the model, the uncertainty in historical production data only affects future mixing ratios through the emission factors. The production data used in the model is only the anthropogenic production. Natural emissions of CH_3Br and CH_3Cl are derived from the constant background mixing ratios and the lifetimes.

The 2008 bank sizes of CFC-11, CFC-12, CFC-115, HCFC-22, HCFC-141b, HCFC-142b, Halon-1211, Halon-1301 are from TEAP (2009). Banks for CFC-113, CFC-114 and Halon-2402 are estimated in WMO (2011). For the other ODSs, CCl_4 , CH_3CCl_3 , Halon-1202, CH_3Br , and CH_3Cl , banks are expected to be small, so it is assumed that emissions are equal to production and that the bank is thus zero. The fraction of the bank emitted annually is calculated in the model from emissions derived from observed mixing ratios and bank sizes averaged over 10 yr (1999–2008). Based on an uncertainty analysis from TEAP/RTOC (2011), an uncertainty of ± 10 % (1σ) is applied

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to the bank sizes of 2008 for all ODS used mainly as refrigerants and an uncertainty of $\pm 20\%$ (1σ) is applied to the bank sizes of the other ODSs. These uncertainties also affect the emission factors, since these factors are calculated as the fraction of the annual emission over the bank. In addition, an extra uncertainty of $\pm 10\%$ is applied to the emission factors of all ODSs used mainly as refrigerant (TEAP/RTOC, 2011) and $\pm 20\%$ for the other ODSs. In the Monte Carlo simulations the emission factor is limited in range from 0 to 1. Uncertainties in bank sizes are assumed to be uncorrelated. Sensitivity tests show that future mixing ratios and EESC levels are not very sensitive to the exact magnitude of the uncertainty in the banks (see Sect. 7).

An uncertainty is also applied to the factor (F_{surf}) which relates the average atmospheric mixing ratio to the corresponding value at the surface. A value of 1.07 is used for all species, except CH_3Br . For CH_3Br a value of 1.16 is used based on Yvon-Lewis et al. (2009). An uncertainty of $\pm 2\%$ (1σ) is assumed for F_{surf} for all species, and it is assumed to be 100 % correlated among species.

The uncertainties in the mean age-of-air and the factor α are from Newman et al. (2007). In the Monte Carlo simulations, the fractional release value is limited in range from 0 to 1 even if the error bars extended past these limits to unphysical values.

Probably the most accurately known quantities used in the model are the observed mixing ratios, taken from WMO (2011). The global and annual average mixing ratios are based on observations by the AGAGE (Prinn et al., 2000) and NOAA networks (Montzka et al., 1999). In the model, the mixing ratios are used to derive top-down emissions and as starting point for the calculation of future mixing ratios. An uncertainty of ± 0.1 ppt (1σ) is assumed for the annual average values. Since the observations are always made in the same way, they will be correlated in time. A time correlation of 0.9 is therefore assumed between successive years. As is shown in Sect. 3.3 the effect of the uncertainty in the observed mixing ratios on future EESC levels is small, so this assumption about the magnitude of the uncertainty is not very important compared with other uncertainties.

If a top-down derived emission for a year becomes negative in a Monte Carlo simulation it is set to zero. This artificially adds emissions to the system and thereby affects the model results, but simulations show that negative emissions occur seldom (less than 0.02 % of the time for most ODSs, but 0.4 % of the time for CFC-113).

Monte Carlo runs are performed with Latin Hypercube Sampling (McKay et al., 1979), which reduces the number of model evaluations significantly, without compromising the accuracy of the results. The statistics that are calculated are the 2.5, 50 and 97.5 percentiles for the production, bank sizes, emissions, mixing ratios, emission fraction for all species as well as for EESC, radiative forcing, and ODP- end GWP-weighted emissions. Uncertainties are presented for the 95 % confidence interval (CI). The same statistics are calculated for the year EESC returns to pre 1980-levels and the integrated EESC above the 1980-level for mid-latitude and Antarctic conditions.

3 Uncertainties in mixing ratios and EESC

We will first focus on the effects of the new lifetimes from SPARC (2013) on mixing ratios and EESC compared with results obtained using the lifetimes from WMO (2011). Then the effects of uncertainties in lifetimes and other parameters on mixing ratios and EESC will be discussed, followed by the effects of the uncertainties on the radiative forcing of the ODSs and on ODPs and GWPs.

3.1 Lifetimes from SPARC vs. WMO

For EESC, the most relevant differences in lifetimes between SPARC (2013) and WMO (2011) are for CFC-11 (+16 %), CCl₄ (+15 %), Halon-1301 (+11 %) and CFC-113 (+9 %). For these species the newer SPARC (2013) lifetimes are all larger than the older ones from WMO (2011). Other relatively large differences are for CFC-115 (−47 %), Halon-1202 (−14 %), Halon-2402 (+40 %), and CH₃Cl (−10 %), but the contribution from anthropogenic emissions of these species to the EESC levels is small

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(currently about 2 % of total EESC). The mixing ratios of the ODSs based on both sets of lifetimes from 1960 to 2100 are shown in Fig. 1. Using the lifetimes from SPARC (2013) instead of those from WMO (2011) results in an increase of 14 ppt (11 %) in the mixing ratio of CFC-11 by 2050, 3.5 ppt (12 %) for CCl_4 , 0.1 ppt (5 %) for Halon-1301, 2.1 ppt (4 %) for CFC-113, 1.2 ppt for HCFC-22 (2 %), and 3.0 ppt for CFC-12 (0.8 %). The increase in mixing ratio for CCl_4 is a result of a slower decrease of the atmospheric burden because of the lower loss rate (longer lifetime) which is in part (about a third) compensated by the smaller future emissions. As discussed in Sect. 2.2 the future emissions of CCl_4 are based on an extrapolation of historic emissions derived from observed mixing ratios and therefore depend on its lifetime.

The effects of the SPARC (2013) vs. WMO (2011) lifetimes on the EESC levels for mid-latitude and Antarctic conditions are shown in Fig. 2. The EESC levels based on the SPARC (2013) lifetimes are only slightly higher than those based on WMO (2011). The year EESC returns to pre-1980-levels for mid-latitude conditions is 2046 based on lifetimes from WMO (2011) and about 2 yr later, 2048, based on lifetimes from SPARC (2013). The corresponding years for Antarctic spring conditions are 2071 for WMO (2011) and about 4 yr later, 2075, for SPARC (2013). The change in integrated EESC above the 1980-level is also small, about 2 % for mid-latitude and about 3 % for Antarctic conditions. The values calculated here with the lifetimes from WMO (2011) differ slightly from those reported in WMO (2011) because of the use of the age-of-air spectrum in the calculations instead of a single mean age-of-air.

3.2 Effects of uncertainties in lifetimes

Apart from a new set of lifetimes, the SPARC assessment also reported uncertainty ranges for these lifetimes. The effects of these uncertainties on the mixing ratios are shown in Fig. 3. It is clear that the uncertainty ranges yields much larger variations in future mixing ratios than do the absolute differences in lifetimes between WMO (2011) and SPARC (2013). For example, the uncertainty in the lifetimes yields a range in mixing ratio of CFC-11 of about ± 35 ppt for the possible range in lifetime and ± 19 ppt

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for the most likely range by 2050, compared with a difference of 14 ppt resulting from the absolute difference in the lifetime of CFC-11 between SPARC (2013) and WMO (2011). For CFC-12, where there is almost no effect from the difference in lifetime between SPARC (2013) and WMO (2011), and the difference in mixing ratio is 3 ppt in 2050, the mixing ratio uncertainty is about ± 48 ppt and about ± 24 ppt in 2050 for the possible and most likely ranges, respectively, in lifetime.

The effects of the ODS lifetime uncertainties on EESC levels is shown in Fig. 4, while in Table 3 the effect of the lifetime uncertainty of each individual ODSs on the year EESC returns to pre-1980 levels is presented. In the baseline scenario, without taking uncertainties into account, EESC returns to pre-1980 levels in 2048 for mid-latitude conditions and in 2075 for Antarctic spring conditions. The individual ODSs that are responsible for the largest uncertainty in the year EESC returns to pre-1980 levels are CFC-11 and Halon-1211, followed by CFC-12, CCl_4 , and CH_3Br . For Antarctic conditions CFC-113 and Halon-1301 are also important due to their relatively long lifetimes. If uncertainties in lifetimes are applied to several ODSs simultaneously, the correlations between uncertainties become important. Correlation is applied to two groups, ODSs that are mainly destroyed by photolysis in the stratosphere and ODSs that are mainly destroyed by reactions with the OH radical in the troposphere. Without correlation the total uncertainty range in EESC and in the year EESC returns to pre-1980 levels is only slightly larger than the largest contribution of the individual ODSs. If the uncertainties are fully correlated the uncertainty range in the year of return is approximately twice as large as when fully uncorrelated.

The species mainly removed in the stratosphere and those mainly removed in the troposphere have similar contributions to the total uncertainty in EESC and in terms of year of return to pre-1980 levels for mid-latitude conditions (possible range of about -5 to $+9$ yr), with the default correlation coefficient of 0.9. For Antarctic conditions the species mainly removed in the stratosphere are the larger contributor to the uncertainty range of -12 to $+24$ yr (possible range), due to their longer lifetimes.

Applying uncertainties to the lifetimes of all ODSs, with a correlation coefficient of 0.9, yields a possible range in the year of return to pre-1980 levels of –8 to +13 yr and a most likely range of –6 to +10 yr for mid latitude conditions. For Antarctic conditions the possible range is –13 to +26 yr and the most likely range is –9 to +14 yr.

The uncertainty in the lifetime of CH₃Cl is not included in the uncertainty analysis since the emissions of CH₃Cl are derived from the observed mixing ratios (and the lifetime) and are assumed to remain constant in the scenario from 2009 onward.

3.3 Overall uncertainty analysis

Apart from the uncertainties in lifetimes, uncertainties in other parameters also affect future mixing ratios and EESC levels. In Fig. 5 the effect of all uncertainties (Table 2) on the normalized EESC levels from 1960 to 2100 is shown. The EESC levels are normalized (1980 level set to 1) in Fig. 5 to avoid a large range in EESC levels in 1980 resulting from uncertainties in the bromine efficiency factor α , in the mean age-of-air, and in the fractional release values. The uncertainty in the mean age-of-air represents the uncertainty in the age-of-air that is considered to be representative for mid-latitude and Antarctic conditions. An alternative approach would have been to calculate the EESC for a particular age and ascribe zero uncertainty to that age; however, we have opted for this approach to account for some uncertainty in the choice of the “best” representative age of the mid-latitude and Antarctic springtime stratosphere. By normalizing, the absolute differences in EESC due strictly to the magnitude of the fractional release values are removed, but there remain impacts of relative gas-to-gas fractional release changes as well as impacts of the differences in the temporal relationship between the tropospheric mixing ratios and EESC at the time of crossing the 1980 level. Also, by normalizing EESC, the year of return to pre-1980 levels can be directly inferred graphically.

The effects of uncertainties of the various parameters on the year EESC returns to pre-1980 levels are shown in Fig. 6 and Table S1 (Supplement). Table S1 also contains data on the integrated EESC above the 1980 level. The largest uncertainty ranges in

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the year of return are caused by the uncertainties in the lifetimes, followed by the uncertainty in the mean age-of-air, with smaller contributions from the uncertainties in the fractional release values and bromine efficiency factor α . The effects of the uncertainty in the production, bank of 2008, and emission factors is small. The reason these last three factors are of minor importance is that the total amount of projected production and anthropogenic ODSs present in existing applications is much smaller than the total atmospheric burden of these ODSs (see also Sect. 7). The changes in the projected atmospheric concentrations are directly affected by the uncertainties in the lifetimes. The EESC levels are also affected by the uncertainties in the fractional release values, bromine efficiency factor, and mean age-of-air. The uncertainties in the year of return shown in Fig. 6 are in good agreement with those found by Newman et al. (2007). The effects of the uncertainty in the fractional release values discussed here is somewhat larger, but that is caused by the larger assumed uncertainty in these values based on Laube et al. (2013).

Combining all uncertainties, the year EESC returns to pre-1980 levels has a possible range of 2039 to 2064 [−9.8 yr, +15.8 yr] and a most likely range of 2040 to 2061 [−8.6 yr, +12.7 yr] for mid-latitude conditions. For Antarctic spring conditions the possible range is 2061 to 2105 [−16.1 yr, +28.3 yr] and the most likely range is 2064 to 2093 [−12.8 yr, +16.9 yr]. The ranges are not very different from those resulting from the uncertainties in the lifetimes alone, showing again that the uncertainties in the lifetimes dominate the total uncertainty in the EESC curve's shape over time. The large sensitivity results from the small EESC slope around the period of recovery compared with the slope near 1980; however, it should also be noted that because of the slow EESC decay projected for the middle part of this century, the absolute difference in ozone depletion could be rather small and still lead to a large difference in the return time.

To put the results of the uncertainty analysis in perspective they are compared with a zero emissions scenario, a zero production scenario, and a scenario with additional emissions. In a scenario with a cease in production of all (anthropogenic) ODSs starting

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in 2014, the year EESC returns to pre 1980-levels is 2045 for mid-latitudes and 2073 for Antarctic springtime, which is about 3 yr earlier than in the baseline scenario in each case. Similarly, if all anthropogenic ODS emissions were to cease in 2014, the year EESC would return to pre-1980 levels is 2037 for mid-latitude conditions and 2063 for Antarctic conditions, about 12 yr earlier than in the baseline scenario. So, the lower end of the uncertainty range, about 2039 for mid-latitudes and 2061–2064 for Antarctic conditions, corresponds approximately to a zero emissions scenario. With respect to the higher end of the uncertainty range in EESC, this can also be reached by a scenario with the best lifetime estimates but with extra ODS emissions on top of the emissions in the baseline scenario. An additional emission of 4 Mt CFC-11 and 4 Mt CFC-12 in 2015 will increase EESC for mid-latitude and Antarctic conditions to such levels that the years EESC return to pre-1980 levels matches the higher end of the uncertainty range. These extra emissions of about 7 Mt CFC-11-eq correspond with 12 times the cumulative HCFC emissions from 2014 to 2050 or about 2 times the cumulative (anthropogenic) emissions of all ODSs from 2014 to 2050.

The uncertainty analyses also yield confidence intervals for the banks and emissions of each ODS. In Fig. S1 (Supplement) the effects of applying uncertainties to all parameters on the banks, emissions, and mixing ratios of the main ODSs are shown. The uncertainty ranges in mixing ratios are almost identical to those of Fig. 3, again demonstrating the key role of the lifetime uncertainty. As discussed in Sect. 7 the uncertainty ranges of HCFC-141b, HCFC-142b, and Halon-1301 are also significantly affected by uncertainties in parameters other than the lifetimes. The ODSs with large banks continuing for several decades are CFC-11, HCFC-22, HCFC-141b, and to a lesser extent, the halons, with CFC-11 having the largest uncertainty in the size of the bank. The uncertainty range in the future emissions is much smaller than in the size of the bank, which is clearly visible, for example, for CFC-11. This different behavior in the size of the bank and future emissions is a result of the way emissions are calculated from the bank and the emission factor. A larger size of the bank corresponds with a smaller emission factor, thereby reducing the effect on future emissions (see also Sect. 7).

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The year 1980 is used here (Table 3 and Fig. 6) and in several WMO ozone assessments as an EESC reference year as one measure of partial ozone recovery. The choice of this particular year and corresponding EESC level is somewhat arbitrary, since observations and model calculations show that ozone depletion occurred before 1980 (WMO, 2011). Using a different year will change the year EESC returns to pre-1980 levels, but will not significantly affect the relative contributions to the uncertainty analysis presented here. For example, the year EESC returns to pre-1975 levels is about 3 decades later than to pre-1980 levels. The EESC slope is smaller at the time of crossing below the 1975 EESC level, which leads to larger uncertainty magnitudes than in the 1980 return case. However, the lifetime uncertainty remains the most important factor to the overall uncertainty.

In Table S1 (Supplement) the effects of the uncertainties in model parameters on EESC are also compared with respect to the change in integrated EESC above the 1980-level. This is a metric commonly used in WMO assessments for comparing different ODS scenarios. It takes into account the time evolution of the ODSs and not only the year EESC-levels drop below the 1980-level.

4 Uncertainties on radiative forcing of ODSs

ODSs do not only deplete the ozone layer they are also strong greenhouse gases (Ramanathan, 1975; Velders et al., 2007). In Fig. 7, the radiative forcing of the ODSs is shown. The uncertainty range in radiative forcing before 2010 is almost completely the result of the applied uncertainty of $\pm 5\%$ (Table 2) in the radiative efficiency of the ODSs (Table 2). From 2020 onward it is almost completely determined by the uncertainty in the lifetimes of the ODSs. The effect of all other uncertainties on the radiative forcing is very small.

The radiative forcing of the ODSs is currently about 0.32 W m^{-2} (WMO, 2011; Montzka et al., 2011a). Taking into account an uncertainty in the radiative efficiency this becomes a range (95 % CI) of 0.30 to 0.34 W m^{-2} . The radiative forcing is ex-

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pected to decrease to 0.17 to 0.23 W m^{-2} in 2050 and to 0.07 to 0.14 W m^{-2} in 2100 (both “possible ranges”). This uncertainty of about $\pm 0.035 \text{ W m}^{-2}$ in 2100 is larger than the difference between the baseline and zero emissions (2014 onward) scenario of 0.03 W m^{-2} in 2050 and 0.006 W m^{-2} in 2100. To put the uncertainty range into perspective it can also be compared with the radiative forcing from, for example, nitrous oxide (N_2O) in the Representative Concentration Pathways (RCPs). In the RCP scenarios the mixing ratio of N_2O increases from about 322 ppb in 2009 (Montzka et al., 2011a) to 342–367 ppb in 2050 (Meinshausen et al., 2011). This corresponds to an increase in radiative forcing from 0.17 W m^{-2} in 2009 to 0.23 – 0.31 W m^{-2} in 2050. So even though ODS concentrations are projected to decline substantially over the rest of this century, the uncertainty in their forcing (about 0.035 W m^{-2}) is significant when compared with N_2O forcing changes (0.06 – 0.14 W m^{-2} in 2050) and other non- CO_2 greenhouse gases.

5 Ozone depletion potentials

A change in the lifetimes of the ODSs also affects ODPs. ODPs are indices that provide a measure to compare the ability of ODSs to destroy stratospheric ozone relative to that of CFC-11 (Wuebbles, 1983; Fisher et al., 1990). ODPs can be calculated using atmospheric chemistry-transport models, but can also be estimated by a semi-empirical approach proposed by Solomon et al. (1992). The semi-empirical ODP is defined as:

$$\text{ODP}_i = (\alpha n_{\text{Br},i} + n_{\text{Cl},i}) \frac{f_i}{f_{\text{CFC-11}}} \frac{\tau_i}{\tau_{\text{CFC-11}}} \frac{M_{\text{CFC-11}}}{M_i} \frac{1}{3} \quad (4)$$

where M_i the molecular weight of species i (in g mol^{-1}), f_i the fractional release value (typically taken for an age-of-air of 3 yr for mid-latitude conditions), α the bromine efficiency factor, n_i the number of chlorine or bromine atoms, and τ_i the lifetime. The

fractional uncertainty is thus defined as the square root of the following:

$$\left(\frac{\sigma_{\text{ODP}_i}}{\text{ODP}_i}\right)^2 \approx \left(\frac{\sigma_{f_i}}{f_i}\right)^2 + \left(\frac{\sigma_{f_{\text{CFC-11}}}}{f_{\text{CFC-11}}}\right)^2 + \left(\frac{\sigma_{\tau_i}}{\tau_i}\right)^2 + \left(\frac{\sigma_{\tau_{\text{CFC-11}}}}{\tau_{\text{CFC-11}}}\right)^2 - 2 \frac{\sigma_{\tau_i}}{\tau_i} \frac{\sigma_{\tau_{\text{CFC-11}}}}{\tau_{\text{CFC-11}}} \text{cor}(\tau_i, \tau_{\text{CFC-11}}) + \left(\frac{\sigma_{\alpha} n_{\text{Br},i}}{\alpha n_{\text{Br},i} + n_{\text{Cl},i}}\right)^2 \quad (5)$$

where σ is the standard deviation of a quantity and $\text{cor}(\tau_i, \tau_{\text{CFC-11}})$ is the correlation coefficient between the lifetimes of species i and CFC-11. ODPs calculated by different models and some uncertainties in the calculation of ODPs have been discussed before (Fisher et al., 1990; Solomon and Albritton, 1992; Solomon et al., 1992), but to our knowledge, an uncertainty analysis as presented here has not been reported before.

In Table 4 the steady-state semi-empirical ODPs are shown using lifetimes from WMO (2011) and SPARC (2013), the fractional release values from WMO (2011), which are the same as those in Newman et al. (2006) for most species, for a mean age-of-air in the stratosphere of 3 yr, and a value of 60 for the bromine efficiency factor (α) in destroying ozone. Table 4 also contains the uncertainties in the calculated ODPs, based on the uncertainties in the lifetimes, fractional release values, and factor α . The calculated ODPs using the lifetimes from SPARC (2013) are for most species lower, up to 25 %, than when using the lifetimes reported in WMO (2011). These differences are mainly the result of the increase of 16 % in CFC-11 lifetime estimate in SPARC (2013). The two exceptions are the ODPs of CFC-115 which is about 50 % lower and of Halon-2402 which is about 20 % higher than in WMO (2011). The uncertainties (95 % CI) in ODPs are 30 to 35 % for the CFCs, 50 to 70 % for the HCFCs, and 60 to 100 % for the bromine containing species (halons and CH_3Br). The uncertainty in the lifetimes is responsible for roughly half of the total uncertainty for each species. Compared with the values defined in the Montreal Protocol, the new ODPs, based on the lifetimes from SPARC (2013) and fractional release values and α from WMO (2011) result in statistically significant different values for CFC-12, CFC-114, CFC-115 and CCl_4 , under the assumption that the ODPs in the Protocol have zero uncertainty. The change in the

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ODP of HCFC-22 is also statistically significant, but only for the most likely uncertainty range.

Recently, Laube et al. (2013) reported new fractional release values for several ODS (see Sect. 2.1). The use of these values also affects the semi-empirical ODPs as shown in Table 4. The different fractional release values alone have the largest impact on the HCFCs with a decrease by about 30 % for HCFC-22 and HCFC-141b and by about 60 % for HCFC-142b when compared with using WMO (2011) fractional release values. Using both the lifetimes from SPARC (2013) and the fractional release values from Laube et al. (2013) results in small changes in ODPs of most species compared with the values reported in WMO (2011). The ODPs of the HCFCs though, show larger changes (decreases); the ODP of HCFC-22 decreases by 37 %, of HCFC-141b by 40 %, and of HCFC-142b by 64 %. Compared with the ODPs in the Montreal Protocol, the fractional release values from Laube et al. (2013) and the lifetimes from SPARC (2013) results in statistically significant differences for only HCFC-22 and HCFC-142b, again under the assumption that the ODPs of the Montreal have zero uncertainty. The changes in the ODPs of CCl_4 , CH_3CCl_3 , and HCFC-141b are also statistically significant, but only for the most likely uncertainty range.

ODP-weighted emissions can be calculated by multiplying emissions directly with ODPs, but this is not the best approach here for calculating uncertainties in the ODP-weighted emissions. The reason is that the uncertainties in the ODS emissions estimated from atmospheric observations of concentration, and in the ODPs both depend on the uncertainties in the lifetimes. These uncertainties will be anti-correlated to some degree and will thus lead to less uncertainty in the calculation of ODP-weighted emission than if they were uncorrelated. The ODP-weighted emissions (Fig. 8) have therefore been calculated using the box model and Monte Carlo analysis and applying uncertainties to all parameters. The ODP-weighted emissions peaked around 1988 at $1.4 \text{ Mt CFC-11-eq yr}^{-1}$ with a possible range from 1.0 to $2.0 \text{ Mt CFC-11-eq yr}^{-1}$. After this peak the emissions continue to decrease towards the background contributions from natural CH_3Br and CH_3Cl emissions.

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The uncertainties in the ODP-weighted emissions vary from 20 to 40 % for the most likely range and 30 to 60 % for the possible range in uncertainties in lifetimes of ODSs. The uncertainty of 20 to 35 % before about 1990 is dominated by the uncertainty in the ODPs of the CFCs and CCl₄. With large decreases in the emissions of these ODSs in the 1990s, the uncertainty in ODP-weighted emissions increase to 40–60 % because of increasing relative contributions of CH₃Br and CH₃Cl, species with larger uncertainty in ODPs than the CFCs.

6 Global Warming Potentials

For species, such as ODSs and other halocarbons, for which the response of a pulse emission can be described by a single exponential decay function, the GWP can be calculated using the following equation,

$$\text{GWP}_i = \frac{R_i}{R_{\text{CO}_2}} \frac{M_{\text{CO}_2}}{M_i} \frac{1}{I_{\text{CO}_2}} \tau_i \left(1 - e^{-\frac{T_H}{\tau_i}} \right) \quad (6)$$

where R_i is the radiative efficiency of species i (in $\text{W m}^{-2} \text{ppb}^{-1}$), I_{CO_2} the integral of the CO₂ response function over the time horizon, M_i the molecular weight (in g mol^{-1}), T_H the time horizon considered (in yr), and τ_i the lifetime (in yr). The parameters for CO₂ (R_{CO_2} and I_{CO_2}) are taken from IPCC (2007). The fractional uncertainty follows from the partial derivatives of the GWP with respect to the various variables,

$$\left(\frac{\sigma_{\text{GWP}_i}}{\text{GWP}_i} \right)^2 \approx \left(\frac{\sigma_{R_i}}{R_i} \right)^2 + \left(\frac{\sigma_{R_{\text{CO}_2}}}{R_{\text{CO}_2}} \right)^2 + \left(\frac{\sigma_{I_{\text{CO}_2}}}{I_{\text{CO}_2}} \right)^2 + \left(\frac{\sigma_{\tau_i}}{\tau_i} \right)^2 \left(1 - \frac{T_H}{\tau_i} \frac{e^{-\frac{T_H}{\tau_i}}}{\left(1 - e^{-\frac{T_H}{\tau_i}} \right)} \right)^2 \quad (7)$$

where σ is the standard deviation of a quantity. An uncertainty of $\pm 5\%$ (1σ) is used for the radiative efficiency R_i and an uncertainty of $\pm 7.5\%$ (1σ) is used for the absolute GWP of CO_2 ($\sim R_{\text{CO}_2} \cdot I_{\text{CO}_2}$) (Table 2).

The GWPs and their uncertainties are shown in Table 5. The GWPs calculated using the lifetimes from SPARC (2013) are within $\pm 5\%$ from those based on the lifetimes from WMO (2011) for most species. Larger differences are seen for the GWP (100 yr) of CFC-11 ($+11\%$), CCl_4 ($+14\%$), and some halons and for the GWP ($T_H = 500$ yr) for several species.

The uncertainty in the GWP depends both on the lifetime and time horizon (see last term in Eq. 7). In general, the longer the lifetime the smaller the uncertainty in GWP and the longer the time horizon the larger the uncertainty. For example, if the time horizon is much smaller than the lifetime, the amount of the emission pulse that remains in the atmosphere over the time horizon is independent of the lifetime. On the other hand, the integrated radiative effect of a pulse becomes linearly dependent on the lifetime for time horizons much longer than the lifetime. The possible uncertainty range in the GWP of CFC-11, for example, more than doubles going from a time horizon of 20 to 500 yr.

The average GWP uncertainty (95 % CI) is about $\pm 24\%$, $\pm 31\%$, and $\pm 36\%$ for a 20, 100, and 500 yr time horizon, respectively, but there is a large spread in the uncertainties among the species depending on the uncertainty in the lifetime and the value of the lifetime, itself. The uncertainty in the lifetime is the major contributor for most species, especially for 100 and 500 yr time horizons.

IPCC (2001, 1996, 2007) quotes an uncertainty in GWPs of $\pm 35\%$, but no reference is given how this value is derived and no confidence interval is specified. This value is probably based on a statement in IPCC (1995) that “suggests uncertainties of less than $\pm 35\%$ ” based on a simple calculation. In IPCC (2007) the $\pm 35\%$ is again quoted without a reference, but this time a 90 % CI is assigned to it. If we assume that the $\pm 35\%$ is for a 95 % CI, it agrees with the average uncertainty of $\pm 31\%$ and $\pm 36\%$ derived here for a time horizon of 100 and 500 yr, respectively. The uncertainty in the IPCC

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reports does not agree with the average uncertainty of $\pm 25\%$ for GWPs for a 20 yr time horizon and it does also not take into account the large range of uncertainties (for example, 18 to 59 % for GWP ($T_H = 100$ yr)) for different species. The GWPs presented in Table 5 are derived for the current atmosphere composition although changes in the composition will certainly affect GWPs. For example, increases in CO_2 mixing ratio for five years (~ 10 ppm) will increase the GWPs of Table 5 by about 2.5 %.

The GWP-weighted emissions have been calculated in the same way as the ODP-weighted emissions using the box model and Monte Carlo analysis. The GWP-weighted emissions (Fig. 9) peaked around 1988 at $9.7 \text{ Gt CO}_2\text{-eq yr}^{-1}$ with a possible range from 8.1 to $11.8 \text{ Gt CO}_2\text{-eq yr}^{-1}$. These emissions have dropped to 1.3 to $2.3 \text{ Gt CO}_2\text{-eq yr}^{-1}$ by 2013 and are projected to continue to decrease. The uncertainty in the GWP-weighted emissions is about 20 % up to 2000 as it is dominated by the contribution from CFC-12. After 2000 the uncertainty increases to about 30 % since the emissions of CFC-12 decrease and the uncertainty is dominated by contributions from HCFC-22.

7 Discussion and conclusions

The magnitude of the uncertainties in the parameters used in the Monte Carlo analysis (Table 2) are based on statistical analysis or on expert judgment and are themselves uncertain. For most parameters, the magnitude of the uncertainty is not very relevant for the outcome of the overall uncertainty analysis. For example, the uncertainty in the production data, bank sizes of 2008, emission factors, observed mixing ratios, and surface factor (F_{surf}) are not very important for future mixing ratios and EESC levels, because the uncertainty in the lifetimes dominate. For some species, however, the uncertainty in the size of the bank of 2008 is relevant for mixing ratio projections. The uncertainty in the future mixing ratios of Halon-1301 are determined roughly equally by the uncertainty in the lifetime and in the size of the bank of 2008, which is a result of the relatively small uncertainty in the lifetime of 9 % compared with an uncertainty in the

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size of the bank of 20 %. For HCFC-141b and HCFC-142b the uncertainty in the size of bank of 2008 makes up about 20 % of the total uncertainty in the mixing ratios in 2050. To understand the effects of the uncertainty in the size of the bank on future mixing ratios it is important to realize how the size of the bank of 2008 is used in the model.

As explained in Sect. 2.1 the size of the bank of 2008 is not only a key starting point for the calculation of future emissions and mixing ratios, it also determines, together with the historic production data and emissions derived from observed mixing ratios, the emission factor for future emissions. Therefore, an increase of the size of the bank of 2008 results in increased future emissions and mixing ratios, but these increases as a result of the larger size of the bank are significantly reduced by a decrease in the emission factor. For HCFCs future emissions not only originate from the bank of 2008, but also from future production. Future cumulative HCFC production in the baseline scenario from WMO (2011) is 2 to 5 times larger than the size of the bank of 2008. This future production is also affected by the change in emission fraction as a result of the uncertainty in the size of the bank. The overall effect of an increase in the size of the bank of 2008 is a decrease in emissions for 10 to 20 yr, followed by an increase afterwards. The turnover point is determined by ratio of the future production and the size of the bank as well as by the emission fraction and thus by the types of applications the ODS is used in.

The magnitude of the uncertainty of the size of bank of CFCs hardly affects the total uncertainty in future mixing ratios, since for these species the size of the bank in 2008 are relatively small compared with total atmospheric burden of the ODSs and production of CFCs has almost completely ceased globally. Of the CFCs, CFC-11 has the largest bank of about 1420 kt in 2008, but this is still a factor of four smaller than its total atmospheric burden in that year. The atmospheric burden of CFC-12 is even about 30 times larger than the size of the bank of 2008.

The uncertainty in the size of the banks of CFC-115 and Halon-2402 are also relevant for the total uncertainty in future mixing ratios (contributing 20 and 10 %, respectively), but these ODSs contribute little to total EESC.

Future production is a quantity that is also relevant for the future mixing ratios of some ODSs. The production of the CFCs, halons, and CH_3CCl_3 has almost ceased globally, but significant production continues for HCFCs and to a lesser extent CH_3Br . The future ODS production in this study is taken from the baseline scenario of WMO (2011) and is assumed to be characterized by a $\pm 5\%$ uncertainty. To put the uncertainty in future mixing ratios of the HCFCs in perspective, it can be compared with a scenario with a cease in production of HCFCs from 2014 onward. In such a scenario the mixing ratios of HCFC-22, HCFC-141b, and HCFC-142b are reduced by 45, 10, and 8 ppt, respectively, in 2050, compared to the baseline scenario. These decreases in mixing ratios are comparable to the differences between the lower end of the uncertainty range and the baseline scenario of 35, 10, and 6 ppt, for HCFC-22, HCFC-141b, and HCFC-142b, respectively, in 2050, applying uncertainties to all parameters. This shows that the effect of the future production of HCFCs results in a change in mixing ratios in 2050 that is comparable to the uncertainty in mixing ratios resulting from the Monte Carlo uncertainty analysis. The effects of the future production of HCFCs on EESC levels is limited, increasing the year EESC returns to pre-1980 levels by 1–2 yr for mid-latitude conditions.

Changes in the mixing ratios of other species – methane, nitrous oxide, and very short-lived species (VSLS), and changes in temperature and atmospheric circulation due to climate change will also affect future ozone levels. Changes in circulation may affect the recovery of the ozone layer by changing the age-of-air in the stratosphere as well as through other effects. Overall, climate change effects are currently expected to accelerate the recovery of the ozone layer at mid-latitudes by a decade or more, but delay it in the Antarctic (WMO, 2011). These aspects have not been considered here, since the focus was on changes and uncertainties in long-lived chlorine and bromine containing species that are controlled by the Montreal Protocol.

Through a comprehensive uncertainty analysis of future mixing ratios of ODSs and EESC levels, we have shown that the uncertainty in the lifetimes of the ODSs is the dominant term. Projections of the recovery of the ozone layer from depletion by ODSs

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depends strongly on knowledge of these lifetime estimates and uncertainties. The year EESC returns to pre-1980 levels is 2048 for mid-latitude conditions and 2075 for Antarctic conditions, based on a baseline scenario for ODS emissions and the lifetimes from the SPARC (2013). Taking all uncertainties in the model parameters into account yields a range for year EESC returns to pre-1980 levels ranges from 2039 to 2064 (95 % CI) for mid-latitude conditions and 2061 to 2105 for Antarctic conditions. Since the current atmospheric burden of CFCs is much larger than the amounts present in existing equipment or still being produced, future mixing ratios and EESC levels strongly depend on the lifetimes and their uncertainties. The lower end of the uncertainty range in EESC is comparable with the EESC levels resulting from a hypothetical scenario with a cease in anthropogenic ODS emissions in 2014. The upper end of the range corresponds with an extra emission about 7 MtCFC-11-eq in 2015. This is equivalent to about 2 times the cumulative anthropogenic emissions of all ODSs, or about 12 times the cumulative HCFC emissions from 2014 to 2050.

Semi-empirical ODPs calculated using the lifetimes from SPARC (2013) and fractional release values from WMO (2011) (mostly based on Newman et al., 2006) are for most species up to 25 % lower than the data reported in WMO (2011) using the lifetimes from that report, mainly as a result of the increasing estimate of the CFC-11 lifetime of 16 %. The ODP of Halon-2402 increases by 20 % and of CFC-115 decreases by 50 %. The uncertainties (95 % CI) in ODPs are about 30 to 35 % for the CFCs, 50 to 70 % for the HCFCs, and 60 to 100 % for the bromine containing species (halons and CH₃Br). Compared with the values defined in the Montreal Protocol, the lifetimes from SPARC (2013) and fractional release values and α from WMO (2011) result in statistically significant differences in the ODPs of CFC-12, CFC-114, CFC-115 and CCl₄, under the assumption that the ODPs in the Montreal have zero uncertainty.

ODSs also contribute to the radiative forcing of climate. Their radiative forcing currently ranges from 0.30 to 0.34 W m⁻² (95 % CI) and is expected to decrease to 0.17 to 0.23 W m⁻² in 2050 and to 0.07 to 0.14 W m⁻² in 2100. The GWPs calculated using the lifetimes from SPARC (2013) are within $\pm 5\%$ from those based on the lifetimes

from WMO (2011) for most species. Larger differences are calculated for the GWPs (100 yr) of CFC-11 (+11 %) and CCl₄ (+14 %). The average uncertainty (95 % CI) for the GWPs of the ODSs is about ±24 %, ±31 %, and ±36 % for a 20, 100, and 500 yr time horizon, respectively. The typical uncertainty of ±35 % reported by IPCC for GWPs

- 5 agrees with the uncertainties derived here in general, but does not take into account the large spread in uncertainties among the species.

Supplementary material related to this article is available online at
[http://www.atmos-chem-phys-discuss.net/13/28017/2013/](http://www.atmos-chem-phys-discuss.net/13/28017/2013/acpd-13-28017-2013-supplement.pdf)
[acpd-13-28017-2013-supplement.pdf](http://www.atmos-chem-phys-discuss.net/13/28017/2013/acpd-13-28017-2013-supplement.pdf).

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Table 1. Lifetimes and uncertainties of ODSs.

	Steady-state lifetime (yr)		Uncertainty in lifetime (1σ) ^c	
	WMO (2011)	SPARC (2013)	Possible	Most likely
CFC-11	45	52	±22 %	±11 %
CFC-12	100	102	±15 %	±8 %
CFC-113	85	93	±17 %	±7 %
CFC-114	190	189	±12 %	
CFC-115	1020	540	±17 %	
CCl ₄	26 ^a	30 ^a	±17 %	±12 %
CH ₃ CCl ₃	5.0 ^a	4.8 ^a	±3 %	
HCFC-22	11.9	12	±16 %	
HCFC-141b	9.2	9.4	±15 %	
HCFC-142b	17.2	18	±14 %	
Halon-1211	16	16	±29 %	
Halon-1202	2.9	2.5	±33 %	
Halon-1301	65	72	±13 %	±9 %
Halon-2402	20	28	±19 %	
CH ₃ Br	0.75 ^{a, b}	0.7 ^a	±17 %	
CH ₃ Cl	1.0 ^a	0.9 ^a	±18 %	

^a Losses due to oceanic and soil processes are taken into account using values from WMO (2011). The partial lifetime for CCl₄ is 44 yr for atmospheric loss and 94 yr for oceanic loss. The partial lifetime for CH₃CCl₃ is 5.0 yr for atmospheric loss and 94 yr for oceanic loss. The partial lifetime for CH₃Br is 1.5 yr for atmospheric loss, 2.2–2.4 yr for oceanic loss and 3.3–3.4 yr for soil loss. The partial lifetime for CH₃Cl is 1.3 yr for atmospheric loss and 3 yr for oceanic and soil loss.

^b In WMO (2011) a best estimate lifetime for CH₃Br of 0.8 yr is reported, but in the scenarios calculations a value of 0.75 yr is used to be consistent with earlier emission estimates.

^c Uncertainty in only the atmospheric loss rate (inverse of the lifetime) from SPARC (2013) is taken into account. This is relevant for CCl₄, for which the uncertainty could change if the uncertainty in the partial lifetime due to oceanic loss (82–191 yr (WMO, 2011)) would be taken into account.

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J. S. Daniel**Table 2.** Parameters and uncertainties used in the model calculations.

	Value	Uncertainty (1σ)	Note
Lifetimes through oxygen photolysis	see Table 1	see Table 1, $\gamma = 0.9$	d
Lifetimes through OH abundance	see Table 1	see Table 1, $\gamma = 0.9$	e
Production data	(WMO, 2011)	$\pm 5\%$	f
Bank size of 2008	(WMO, 2011) ^a	$\pm 10\%$ for CFC-12, CFC-115, HCFC-22, $\pm 20\%$ for other ODSs	g
Emission factor (E_f)	Calculated	$\pm 10\%$ for CFC-12, CFC-115, HCFC-22, $\pm 20\%$ for other ODSs	g
Alpha mid-latitudes	60	$\pm 25\%$	h, i
Alpha Antarctic	65	$\pm 25\%$	h, i
Mean age-of-air mid-latitudes	3.0	± 0.3 yr	i
Mean age-of-air Antarctic	5.5	± 0.3 yr	i
Fractional release	(Newman et al., 2006) ^b	$\pm 20\%$ for HCFCs, $\pm 10\%$ for other ODSs	j
Observed mixing ratios	–	± 0.1 ppt, $\gamma = 0.9$	k
Surface factor (F_{surf})	1.07 ^c	$\pm 3\%$	l
Radiative efficiency	(WMO, 2011)	$\pm 5\%$	m
Absolute GWP of CO ₂	(IPCC, 2007)	$\pm 7.5\%$	n

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Table 2. Continued.

^a The sizes of the banks of 2008 as used in WMO (2011). The following bank sizes are from bottom-up analysis of TEAP (2009): 1420 kton for CFC-11, 394 kt for CFC-12, 16 kt for CFC-115, 1618 kt for HCFC-22, 941 kt for HCFC-141b, 273 kt for HCFC-142b, 74 kt for Halon-1211, and 47 kt for Halon-1301. The following bank sizes are estimated in WMO (2011): 25 kt for CFC-113, 15 kt for CFC-114, and 11 kt for Halon-2402.

^b Fractional release values depend on the age-of-air. The parameterizations of Newman et al. (2006) are used here, except for HCFC-141b and HCFC-142b. As argued in WMO (2011), the abundances of HCFC-141b and HCFC-142b were small and had large temporal trends at the time the measurements upon which these fractional release values are based were made, leading to large uncertainties. Therefore, the functions describing the age-of-air dependent fractional release values for HCFC-141b has been adjusted to be in agreement with the values used in WMO (2011) for 3 and 5.5 yr mean ages. The functions of HCFC-142b have also been adjusted to be in agreement with the value in WMO (2011) for 3 yr mean age-of-air and follow the shape of the curves of HCFC-141b. The fractional release value for HCFC-142b for 5.5 yr mean age-of-air in Newman et al. (2006) and WMO (2011) are much lower than the values of similar ODSs and has therefore not been used. For 5.5 yr mean age-of-air a fractional release value of 0.65 for HCFC-142b has been used to obtain an age-of-air dependent fractional release curve resembling the one for HCFC-141b. The functions for the fractional release used are for HCFC-141b = $0.020388 \times \text{age}^2 + 0.051503 \times \text{age}$; for HCFC-142b = $0.024739 \times \text{age}^2 - 0.017885 \times \text{age}$.

^c A value of 1.16 is used for CH₃Br, as was done in WMO (2011), based on Yvon-Lewis et al. (2009)

^d The correlation in uncertainty in lifetimes of species that are mainly destroyed in the stratosphere by photolysis (CFCs, CCl₄ and Halon-1301) is assumed to be large. A correlation coefficient (γ) of 0.9 means a 67 : 33 distribution of the random numbers in Eq. (3).

^e The correlation in uncertainty in lifetimes of species that are mainly destroyed in the troposphere by reaction with OH (HCFCs, CH₃CCl₃, Halon-1211, Halon-1202, Halon-2402, CH₃Br, CH₃Cl) is assumed to be large. A correlation coefficient (γ) of 0.9 means a 67 : 33 distribution of the random numbers in Eq. (3). The fact that a significant fraction of Halon-1211 and -2402 is removed in the stratosphere is ignored in the correlation coefficient.

^f The uncertainty in global production data of $\pm 5\%$ (1σ) is assumed. The historic production data is from a small number of companies and officially reported by countries to UNEP (2008).

^g An uncertainty (lower and upper threshold) of 18.2% (interpreted here 2σ) is given for the bank sizes of refrigerants in TEAP/RTOC (2011). An uncertainty (lower and upper threshold) of 26.6% (interpreted here 2σ) is given for the emissions of refrigerants in TEAP/RTOC (2011). In combination with the uncertainty in the banks (see g), this yields an uncertainty in the emission factor of about $\pm 10\%$ (1σ). An uncertainty of $\pm 10\%$ (1σ) is therefore applied to the banks and emission factors of ODSs that are mainly used as refrigerant and for which the bank size is from TEAP (2009). A larger uncertainty of $\pm 20\%$ (1σ) is assumed for the banks and emission factors of ODSs that are used mainly in foams, solvents and fire-extinguishers.

^h Alpha value from WMO (2011).

ⁱ Uncertainty from Newman et al. (2007).

^j Uncertainty from Laube et al. (2013).

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**Table 2.** Continued.

^k An uncertainty if the global and annual average observed mixing ratio of ± 0.1 ppt (1σ) is assumed with time correlation coefficient (γ) of 0.9.

^l An uncertainty of $\pm 3\%$ (1σ) is assumed.

^m An uncertainty of $\pm 5\%$ (1σ) is used, based on a reported uncertainty of $\pm 10\%$ (assumed to be 2σ) by IPCC (2001).

ⁿ An uncertainty of $\pm 7.5\%$ (1σ) is used for the absolute GWP of CO_2 ($\sim R_{\text{CO}_2} \cdot I_{\text{CO}_2}$ in Eq. 7), based on a reported uncertainty of $\pm 15\%$ (assumed to be 2σ) by IPCC (2007).

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J. S. Daniel**Table 4.** Ozone depletion potentials (ODPs) and their uncertainties. The ODPs shown are those included in the Montreal Protocol, the semi-empirical values reported in WMO (2011), and the semi-empirical values calculated here using the lifetimes from SPARC (2013) and fractional release values from WMO (2011) and Laube et al. (2013).

	Montreal Protocol	WMO (2011)	Fractional release WMO	Fractional release Laube		Uncertainties (95 % CI) ^b	
			Lifetimes SPARC	Lifetimes WMO	Lifetimes SPARC	Possible (±)	Most likely (±)
CFC-11	1	1	1	1	1		
CFC-12	1.0	0.82	0.73	0.91	0.81	34 %	30 %
CFC-113	0.8	0.85	0.81	0.87	0.82	34 %	30 %
CFC-114	1.0	0.58	0.50			37 %	30 %
CFC-115	0.6	0.57	0.26			34 %	32 %
CCl ₄	1.1	0.82	0.82	0.83	0.82	34 %	30 %
CH ₃ CCl ₃	0.1	0.16	0.14	0.20	0.17	52 %	36 %
HCFC-22	0.055	0.04	0.034	0.028	0.024	69 %	58 %
HCFC-141b	0.11	0.12	0.102	0.078	0.069	68 %	57 %
HCFC-142b	0.065	0.06	0.057	0.025	0.023	67 %	56 %
Halon-1211	3.0	7.9	6.9	8.9	7.7	90 %	82 %
Halon-1202		2.2	1.7			96 %	88 %
Halon-1301	10.0	15.9	15.2	19.8	19.0	61 %	57 %
Halon-2402	6.0	13.0	15.7			80 %	71 %
CH ₃ Br	0.6	0.66 ^a	0.50			78 %	69 %
CH ₃ Cl		0.02	0.015			62 %	50 %

^a This value was based on a best estimate lifetime of CH₃Br of 0.8 yr.^b In the calculation of the uncertainties a correlation coefficient of 0.9 is used for the lifetimes of CFC-11 and the other CFCs, CCl₄ and Halon-1301.

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Table 5. Global warming potentials (GWPs) and their uncertainties. The GWPs shown are the values reported in WMO (2011) and the values and uncertainties calculated here using the lifetimes from SPARC (2013).

	This work based on						Uncertainties (95 % CI)					
	WMO (2011)			lifetimes SPARC (2013)			Possible range (±)			Most likely range (±)		
	20 yr	100 yr	500 yr	20 yr	100 yr	500 yr	20 yr	100 yr	500 yr	20 yr	100 yr	500 yr
CFC-11	6730	4750	1620	6940	5260	1870	19 %	34 %	47 %	18 %	23 %	28 %
CFC-12	11 000	10 900	5200	11 030	10 990	5300	18 %	22 %	35 %	18 %	19 %	23 %
CFC-113	6540	6130	2690	6610	6390	2930	18 %	23 %	37 %	18 %	19 %	22 %
CFC-114	7890	9180	6330	7900	9170	6310	18 %	19 %	26 %			
CFC-115	5290	7230	9120	5250	6930	7520	18 %	18 %	22 %			
CCl ₄	2700	1,400	435	2830	1590	502	20 %	34 %	38 %	19 %	27 %	29 %
CH ₃ CCl ₃	506	146	45	488	141	43	19 %	19 %	19 %			
HCFC-22	5130	1790	545	5170	1810	550	26 %	36 %	36 %			
HCFC-141b	2240	717	218	2280	733	223	27 %	34 %	34 %			
HCFC-142b	5390	2220	678	5520	2320	709	22 %	32 %	33 %			
Halon-1211	4750	1890	575	4750	1890	575	33 %	59 %	60 %			
Halon-1202												
Halon-1301	8480	7140	2760	8610	7570	3,060	18 %	22 %	31 %	18 %	20 %	25 %
Halon-2402	3680	1640	503	4170	2250	704	21 %	38 %	41 %			
CH ₃ Br	19	5	2	17	5	1	38 %	38 %	38 %			
CH ₃ Cl	45	13	4	41	12	4	39 %	39 %	39 %			

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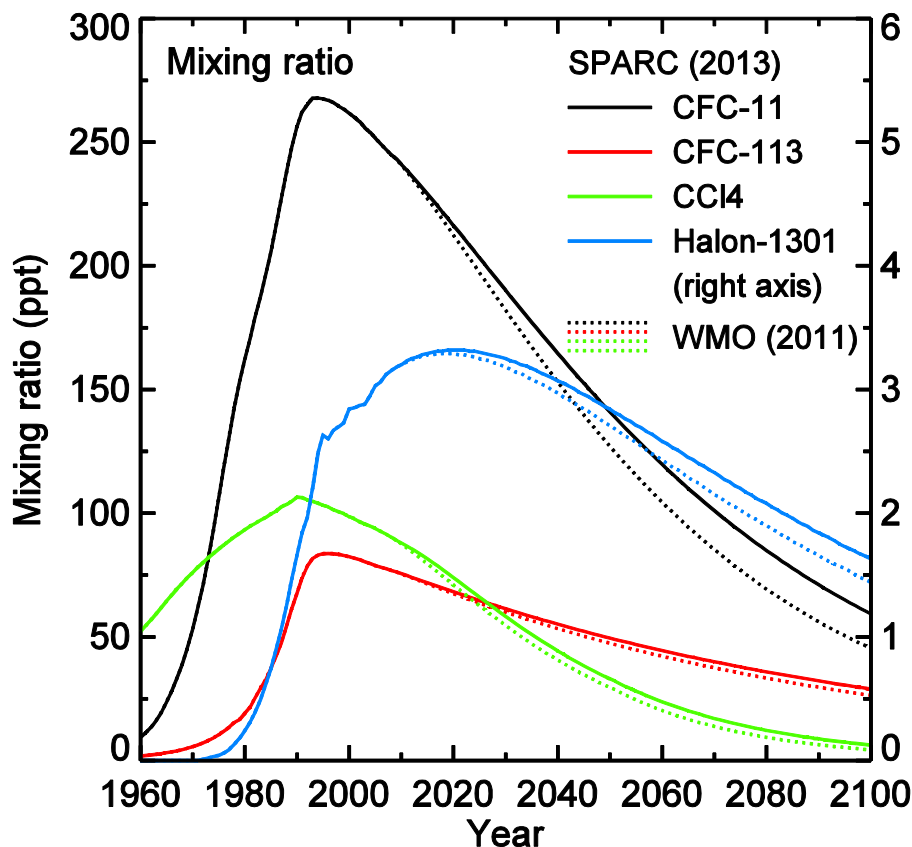


Fig. 1. Mixing ratios based on lifetimes from SPARC (2013) and WMO (2011). The mixing ratios are shown for the ODSs with the for EESC most relevant differences in lifetimes. Before 2010, concentrations are constrained by observations or are fixed from some other information and so the lifetime differences affect calculated emissions but not concentrations.

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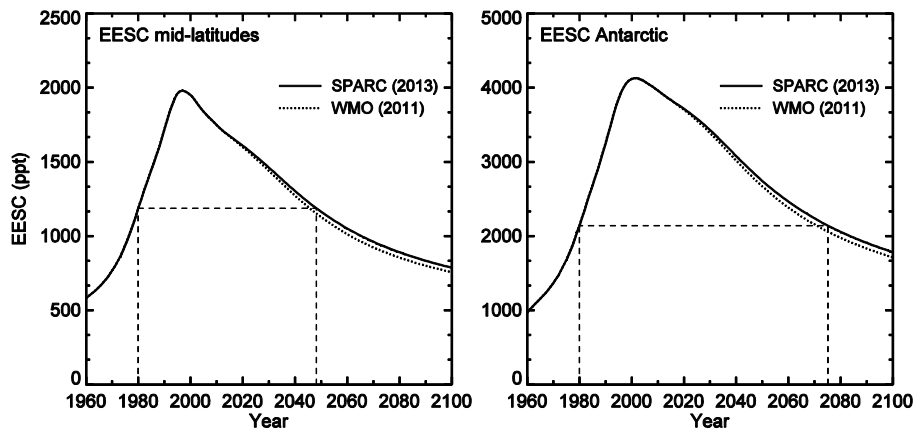


Fig. 2. EESC for mid-latitude (left) and Antarctic (right) conditions based on lifetimes from SPARC (2013) and WMO (2011). The thin dashed lines indicate the 1980 EESC levels and the years the EESC curves cross these levels.

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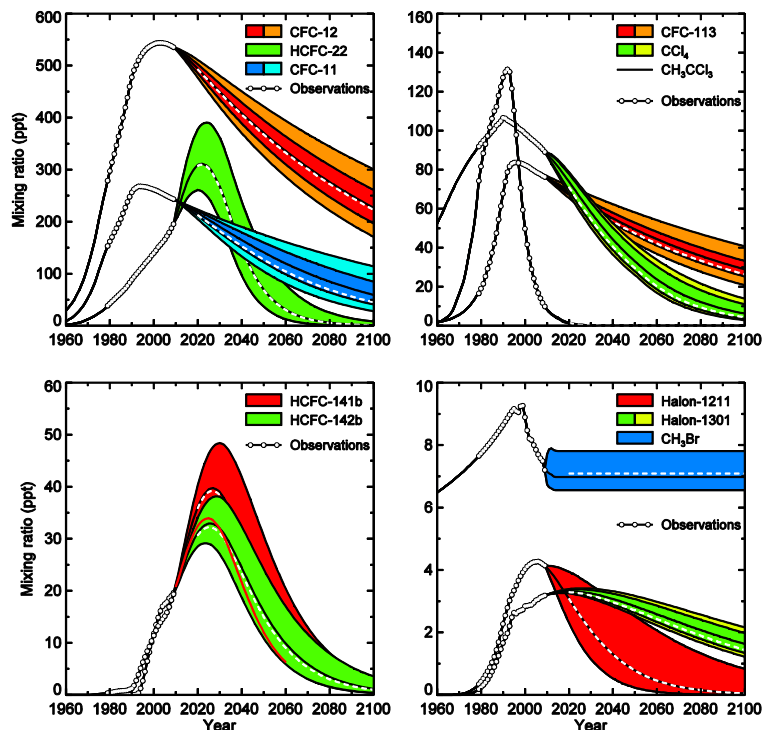
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Fig. 3. Mixing ratios of ODSs from 1960 to 2100 based on lifetimes and their uncertainties from SPARC (2013). Shown are the median values and 95 % CI based on the most likely (dark colors) and possible (light colors) uncertainty ranges in lifetimes. Open circles show the observed mixing ratios. White dashed lines represent the median values of the mixing ratios based on lifetimes from WMO (2011).

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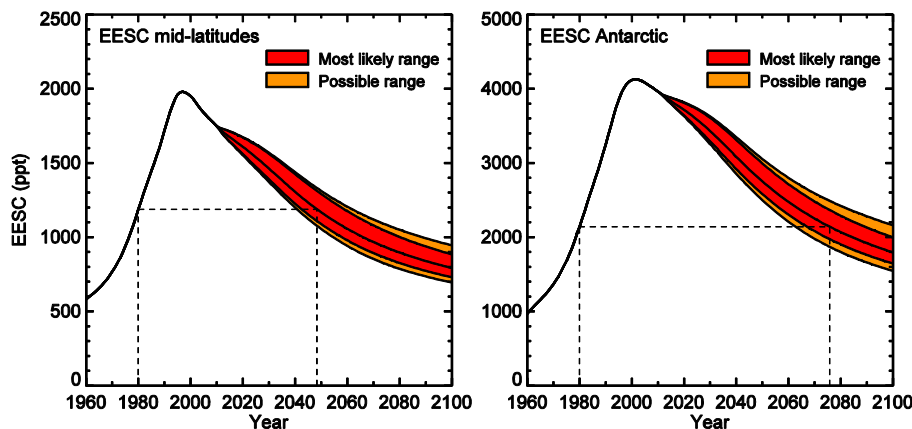
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Fig. 4. EESC from 1960 to 2100 for mid-latitude (left) and Antarctic (right) conditions using the lifetimes from SPARC (2013) and with uncertainties applied to the loss rates (inverse of lifetimes) of all species, except CH_3Cl . A correlation coefficient γ of 0.9 is used for the uncertainties in lifetimes among the ODSs mainly removed in the stratosphere and among those mainly removed in the troposphere. Shown are the median values and 95 % CI based on the possible (orange) and most likely (red) uncertainty ranges in lifetimes. The years EESC return to pre-1980 levels for the median EESC values is indicated with thin dashed lines.

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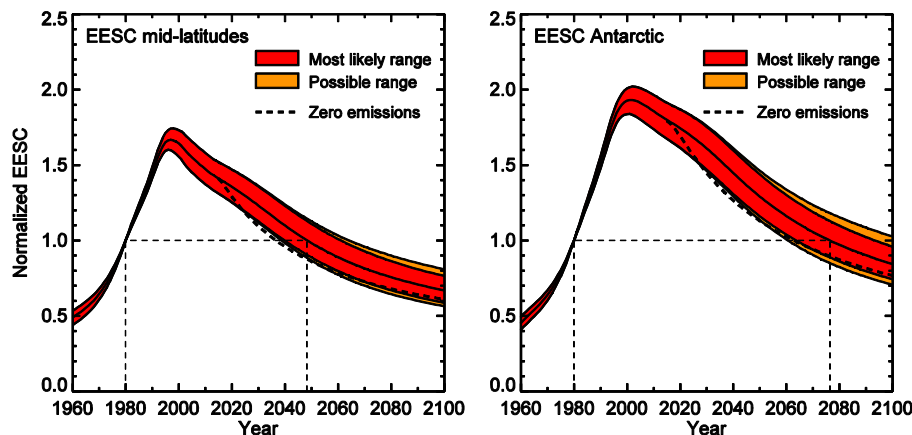
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Fig. 5. Normalized EESC (1980 value set to 1) from 1960 to 2100 for mid-latitude (left) and Antarctic (right) conditions using the lifetimes from SPARC (2013) and with uncertainties applied to all parameters. Shown are the median values and 95 % CI based on the possible (orange) and most likely (red) uncertainty ranges in lifetimes. The years EESC return to pre-1980 levels for the median EESC values is indicated with thin dashed lines. The EESC curves corresponding to the zero emission scenario are shown for reference.

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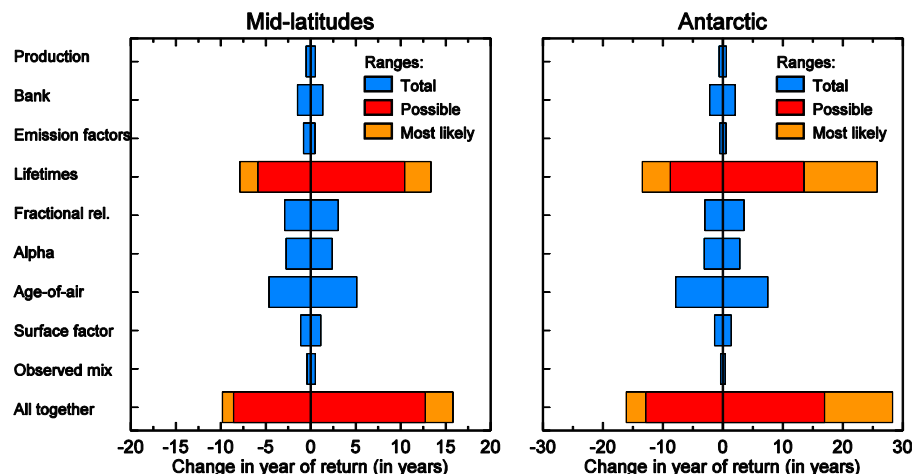
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Fig. 6. Uncertainties (difference from the median, 95 % CI) in the years of return to pre-1980-levels for mid-latitude and Antarctic springtime conditions resulting from uncertainties in model input parameters. The year of return in the base run is 2048 for mid-latitudes and 2075 for Antarctic spring conditions.

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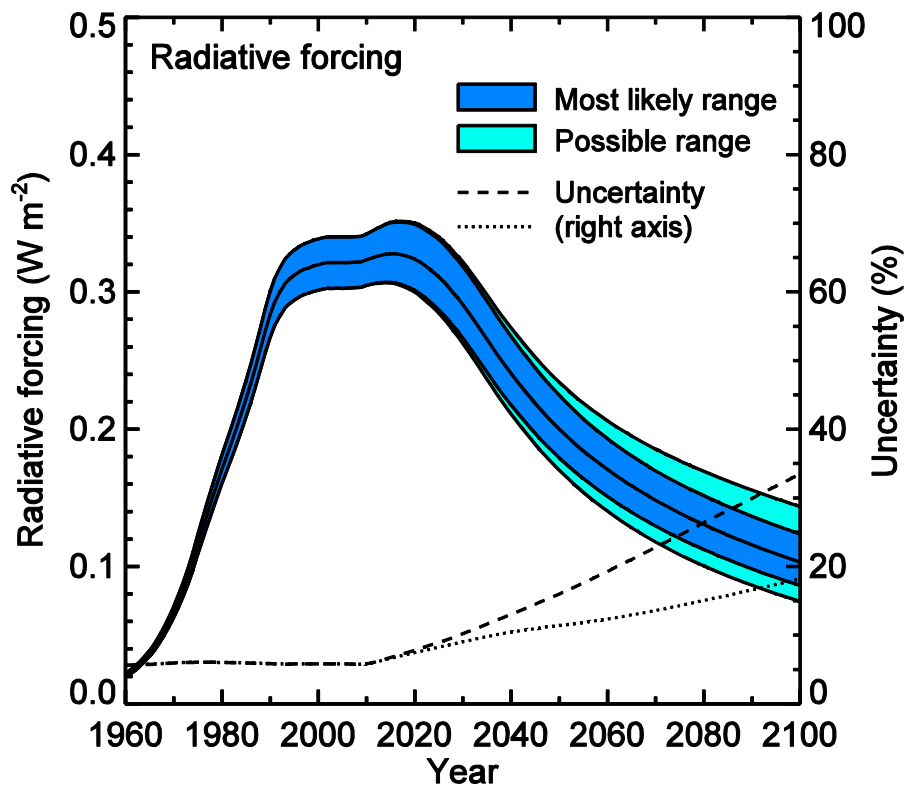


Fig. 7. Radiative forcing of all ODSs from 1960 to 2100 using the lifetimes from SPARC (2013) and with uncertainties applied to all parameters, including the radiative efficiencies. Shown are the median values and 95 % CI based on the possible (orange) and most likely (red) uncertainty ranges in the lifetime contributions to the total uncertainties. The uncertainties (95 % CI) are also shown as percentages of the median for the possible (dashed line) and most likely (dotted line) ranges.

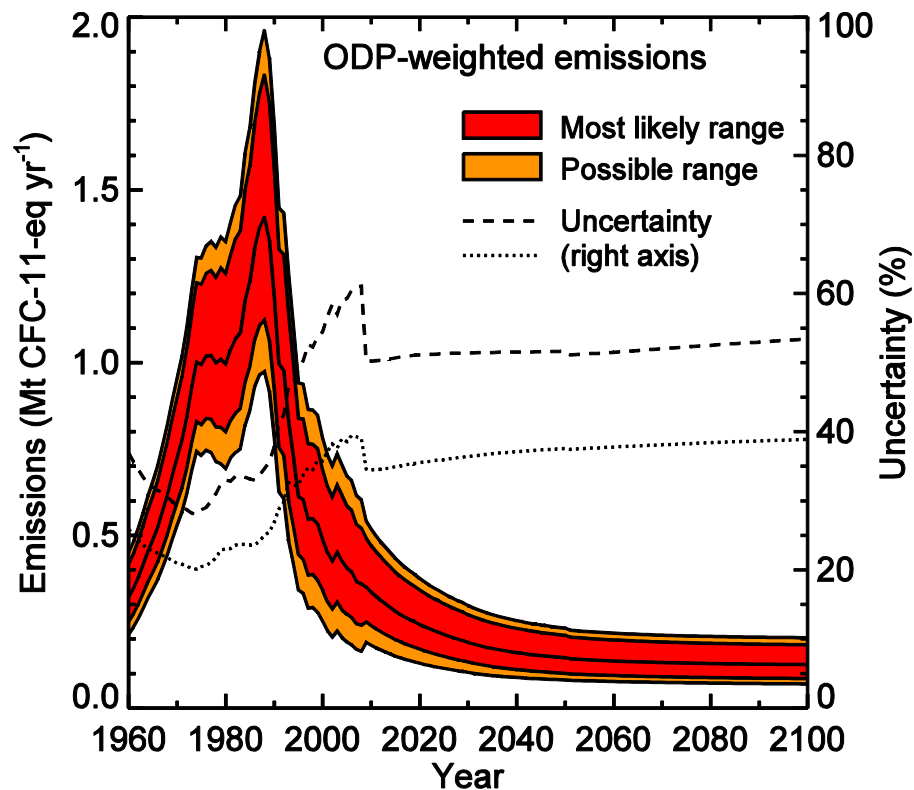


Fig. 8. ODP-weighted emissions from 1960 to 2100 using the lifetimes from SPARC (2013) and with uncertainties applied to all parameters. Shown are the median values and 95 % CI based on the possible (orange) and most likely (red) uncertainty ranges in lifetimes. The uncertainties (95 % CI) are also shown as percentages of the median for the possible (dashed line) and most likely (dotted line) ranges.

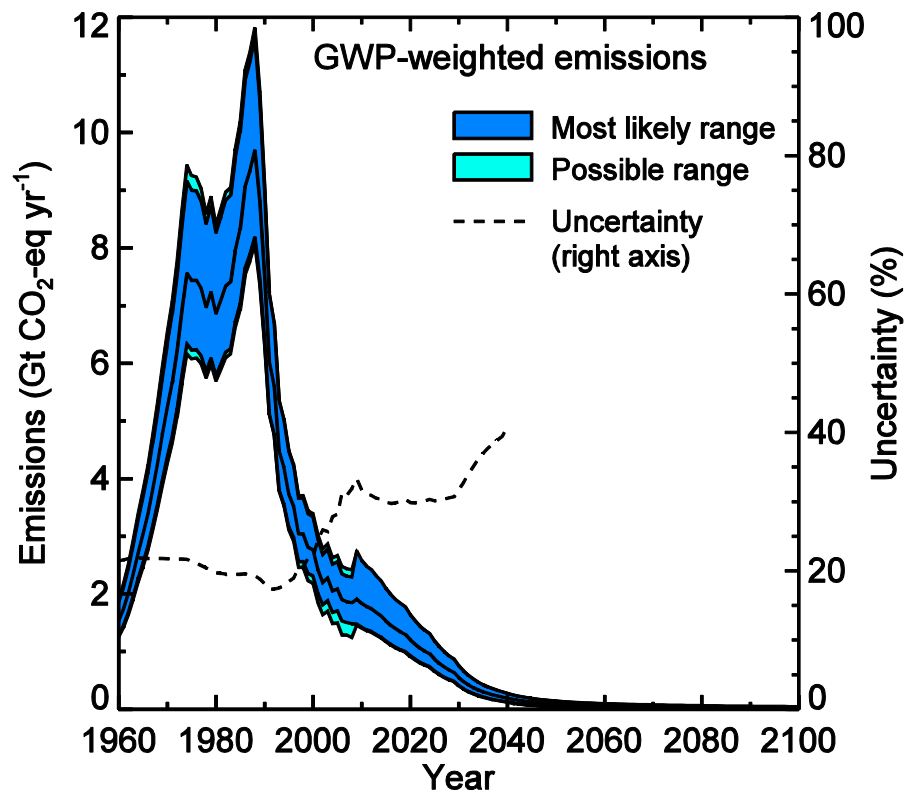


Fig. 9. GWP-weighted emissions from 1960 to 2100 using the lifetimes from SPARC (2013) and with uncertainties applied to all parameters. Shown are the median values and 95 % CI based on the possible (light blue) and most likely (dark blue) uncertainty ranges in lifetimes. The uncertainties (95 % CI) are also shown as percentages (dashed line) until 2040, when the emissions become very small.