**Integrated ozone depletion as a metric for ozone recovery**

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**The Montreal Protocol is successfully protecting the ozone layer. The main halogen gases responsible for stratospheric ozone depletion have been regulated under the Protocol, their combined atmospheric abundances are declining and ozone is increasing in some parts of the atmosphere1. Ozone depletion potentials (ODPs)2,3,4, relative measures of compounds’ abilities to deplete stratospheric ozone, have been a key regulatory component of the Protocol in successfully guiding the phase out in manufacture of the most highly depleting substances. However, this latest, recovery phase in monitoring the success of the Protocol calls for additional metrics. The ‘delay in ozone return’ has been widely used to indicate the effect of different emissions or phase-down strategies, but we argue here that it can sometimes be ambiguous or even of no use. Instead, we propose the use of an integrated ozone depletion (IOD) metric to indicate the impact of any new emission. The IOD measures the time-integrated column ozone depletion and depends only on the emission strength and the whole atmosphere and stratospheric lifetimes of the species considered. It provides a useful complementary metric of the impact of specific emissions of an ODS for both the scientific and policy communities.**

Ozone depletion potentials have been a key metric for the Montreal Protocol, guiding control measures for the protection of the ozone layer. Following the success of the Protocol in the phasing out of potent ozone depleting substances (ODS) and the subsequent decline in the aggregated stratospheric levels of both chlorinated and brominated ODS, which started around the turn of the century, there are now indications of positive trends in ozone, at least in particular locations, so that we are now into a new phase, monitoring the recovery of the ozone layer5,6,7,8,9. Threats to the ozone layer now come from either illegal breaches of the Protocol10,11 or from emissions of unregulated short-lived substances of anthropogenic origin12,13. We believe this new phase calls for a new metric, the integrated ozone depletion, described below. The impact of these new threats has so far been addressed largely by considering for how long they might delay the time for return of column ozone, globally or regionally, to some previous value, often taken to be the 1980 value14,15,16 (although the build-up of anthropogenic ODS in the atmosphere was already well established by then). However, we argue here that return to a particular value is not necessarily the best, or most useful, metric.

Figure 1 is a schematic illustration of possible future ozone trajectories under a range of different future emissions of long and short-lived gases, based on integrations discussed later. The black line in Figure 1 is the anticipated projection of global mean column ozone levels assuming full compliance with the Montreal Protocol; in this example return to the historic threshold value (taken here as ~1960, before which time ODS emissions into the atmosphere were small, and given by the horizontal dashed line) occurs sometime in the mid 2070s. Increasing levels of CO2 will cool the stratosphere and tend to increase global stratospheric ozone concentrations so that ozone will likely exceed the 1960 value later this century. Clearly, the black line will depend on the particular evolution of both the ODS and greenhouse gas (GHG) concentrations. The blue line is the ozone trajectory assuming the same GHG scenario but that some additional long-lived halocarbons, either unregulated or not compliant with the Montreal Protocol, are emitted for a few years and reach the stratosphere, in which case the return to the 1960 value is delayed to the early 2080s.

But is giving the delay in a return date useful? There are several reasons why it is not. If the new (blue) recovery line has a gradient with time which is very close to zero (i.e., close to the horizontal dashed line in Figure 1), then the delay will be long but the actual ozone depletion during the delay may be very small. A key point here is that ozone would not return to its past value in a single instant. Instead, there could be a long period of time in which total column ozone is within the natural variability of past periods, constantly rising above and falling below the chosen threshold value for periods of years to decades16. Furthermore, if the stratosphere is subjected to emissions of a very short-lived halogenated substance, there may be significant ozone depletion of short duration (e.g., the red line in Figure 1) but with no delay in the time for ozone to return to a previous, historic value. Having a metric which provides information along the full pathway to recovery may better reflect total column ozone changes associated with short-lived species. Finally, assessing the time delay before the return date is reached requires a detailed calculation, usually performed using a computationally expensive 3D model (a chemical transport model or a chemistry-climate model). This metric is not simple, requires the detailed calculation for every possible compound and its emission scenario, and would be subject to inter-model variability. For all the above reasons, we argue that a new, complementary metric is required now that we are in this new phase of ozone recovery. The metric should (i) provide more information about the pathway to recovery than the return date, (ii) allow the assessment of the impact of a range of emissions of an ODS and (iii) be accessible without repeated 3D model calculations.

We have explored a metric which satisfies these criteria. We argue that an important measure of the impact of any new emissions for policy is the cumulative additional ozone depletion consequent on those emissions. For the hypothetical ozone depletion by additional emissions of a long-lived ODS in Figure 1, the cumulative additional ozone loss (hereafter, the Integrated Ozone Depletion, IOD) would be the integral between the blue and black lines (the blue shaded area) or, for the hypothetical depletion by a short-lived substance, the integral between the red and black lines (the red shaded area). The time period over which these scenarios differ from one another is controlled by the duration of the additional emission, the lifetime of the species and the subsequent removal of active halogen from the stratosphere. Dividing the IOD by the time period of the perturbation provides some estimate of the annual average ozone loss. Alternatively, IODs could be used to determine the percentage increase in additional ozone loss by comparing the time integrals for this additional loss (e.g., the red and blue shaded areas) with ozone loss that should occur under full compliance with the Protocol until some recovery measure is reached (the yellow shaded area in Figure 1).

We have investigated ozone projections, return dates and the new IOD metric using our chemistry-climate model, UM-UKCA, with a range of integrations representing several different scenarios. A description of the model and all the scenarios used is provided in the Methods; Table 1 lists the scenarios. Some of these integrations look at the impact of new emissions of CFC-11, aimed at addressing the recent emissions detected in contravention of the Montreal Protocol10,11,17. CFC-11 has an atmospheric lifetime of about 55 years, but recent observations have also documented an increase in the atmospheric concentrations of a number of unregulated short-lived halogenated substances (dichloromethane (DCM), chloroform, dichloroethane (DCE), etc)12,13,18 which could also have an impact on the pathway of ozone recovery. Accordingly, we have also explored the impact of a range of hypothetical emissions of gases with shorter atmospheric lifetimes than CFC-11. These integrations were not designed to simulate the impact of any particular gas but instead to explore a range of emissions and lifetimes to allow some general conclusions to be drawn. We have taken CH3CCl3 (methyl chloroform, MCF), CH3Cl and CH3Br as examples, with whole atmosphere lifetimes of 5.0, 0.9 and 0.8 years, respectively1. (Lifetimes taken from the literature and calculated from UM-UKCA simulations are given in Table 2). They have shorter atmospheric lifetimes than CFC-11 but somewhat longer atmospheric lifetimes than many short-lived gases, so any increase in their emissions would likely have a larger, more long-lasting impact than for the shortest-lived gases. We have followed this approach because emissions of DCM, DCE, etc. are poorly known, with projections of future trends even more so. So, in addition to a Base integration, which followed WMO 2018, and the CFC calculations mentioned above, we carried out additional integrations considering a very large range of emissions of MCF, CH3Cl and CH3Br. Together these integrations allow us to study the new metric over a very wide range of conditions.

Figure 2 shows timeseries of global mean, annual mean total column ozone from one selected emission scenario for each of CFC-11, MCF and CH3Br (the evolution of global column ozone for all the scenarios explored in this study is shown in Extended Data Figure 1). The calculated trajectory of recovery for the CFC-11 emission case (Scenario 6, Table 1) resembles the case exemplified by the hatched blue area in Figure 1 where, although a delay is modelled, the magnitude of the depletion during the delay period is comparable with the natural variability of the global mean, which we have estimated as about ±1.5% or about 5 DU, based on Fioletov et al.19 and similar to the model variability seen in our ensemble of integrations. This particular scenario assumes that while part of the produced CFC-11 is emitted directly to the atmosphere, the majority enters a bank and is emitted at a slower rate over the course of the 21st century. For MCF the largest departures from the Base occur, as expected, in the integrations with the largest additional cumulative emissions (1000 Gg yr-1 for 10 years, pale blue curve). Return to the 1960 value occurs in the Base run in about 2075, a return date which is indistinguishable from that in the MCF integrations including these large, additional emissions. This resembles the case illustrated schematically by the red line in Figure 1. This is demonstrated even more clearly for the CH3Br emissions. The huge emissions modelled here (380 Gg yr-1 for 5 years which should be compared with annual anthropogenic emissions1 for quarantine and pre-shipment (QPS) and other anthropogenic uses peaking during the late 1990s at about 50 Gg yr-1) lead to a very substantial mean global ozone depletion of about 20 DU soon after the emissions occur but with absolutely no change in return date. Clearly in this case (and, by extrapolation, the case of large emissions of short duration for other short-lived gases), and despite a substantial ozone loss, there is no difference in return dates for global ozone; the return date is clearly demonstrated in this case not to be a useful metric either for environmental impact or for policy.

A more useful metric would be the time-integrated ozone depletion arising from new emissions, additional to the depletion in the Base run in which full compliance with the Montreal Protocol is assumed. Under clear sky conditions, this additional ozone loss could be related to an increase in UV at the surface (assuming that cloudiness, etc., remains constant) and, using the appropriate action spectrum, to the impact, for example, on erythema or DNA damage, where a 1% decrease in ozone translates roughly into a 1-2% increase in UV dose in those two cases20.

The impact of any halogenated source gas on stratospheric ozone depletion will depend on the total emission of the gas and the fraction of that emission that is transported into and reacts in the stratosphere to liberate inorganic halogens, i.e. the fraction of emitted halogen that is available to take part in stratospheric ozone depletion reactions. This fraction is dependent on a number of complex atmospheric processes but can be calculated simply as the whole atmosphere lifetime (τatmos) of a species divided by its stratospheric lifetime (τstrat) (see Methods for further details). How much of an ODS degrades in the stratosphere is, of course, recognised in the definition of the ODP4.

Consider a long-lived gas like CFC-11 which has no significant tropospheric loss so that all of its emissions will eventually be destroyed chemically in the stratosphere. In that case, for CFC-11 (and other species with negligible tropospheric loss), τatmos/τstrat = 1, and all the chlorine emitted is ultimately available for ozone depletion. In contrast, for shorter lived species which undergo significant destruction in the troposphere (e.g. MCF with tropospheric and stratospheric lifetimes of respectively ~5 years and ~40 years) τatmos/τstrat << 1 and only a fraction of the emitted chlorine is available for stratospheric ozone depletion. The tropospheric lifetimes of very short-lived halocarbons, and therefore the amount of the emission that degrades in the stratosphere, are uncertain with variability arising from the location and season of the emissions and the variability, on many timescales, in atmospheric transport.

Figure 3 shows the integrated ozone depletion (IOD) for the various emission scenarios of the different gases plotted against the total halogen emission in Tg Cl, EEq, multiplied by the ratio of the whole atmosphere lifetime of the particular gas to its stratospheric lifetime, τatmos/τstrat. We have used a bromine efficiency factor of 60 in the calculation of EEq for brominated species, only slightly lower than some recent suggestions21. To a very good approximation, all the data points sit on a straight line with deviations arising from the variability in meteorology inherent in our use of a chemistry-climate model. The IOD in Figure 3 is calculated over the entire period of the integration, although once the halogen emitted from the short-lived gas has been removed from the stratosphere there should be no further depletion. Due to our use of a chemistry-climate model, we did find some contributions to IOD well beyond the expected period of the perturbation. When the IOD was calculated over a perturbation period that depends on the atmospheric lifetime of the species of interest and an estimate of the stratospheric residence time22,23 we obtained very similar results to those in Figure 3, with an identical gradient but a better correlation coefficient (Extended Data Figure 2).

Given the linear relationship shown in Figure 3, we can write

$IOD=K E\_{Eq} \left(\frac{τ\_{atmos}}{τ\_{strat}}\right) $ Equation (1)

where *K* = 100±16 DU years/Tg(Cl). The equation thus allows the impact on global ozone of any emission of a halogenated gas of known lifetimes to be assessed in a simple fashion. The IOD could be compared with the expected integrated ozone depletion arising from full compliance with the Protocol (equivalent to the yellow shaded area in Figure 1, which for our Base calculation is 1440 DU years) to indicate the additional depletion relative to the Base case.

The metric could also be used to provide a very approximate estimate of the annual average ozone depletion arising from the emissions if IOD is divided by the length of the perturbation (or the integration length in the case of very long-lived gases). There is a large range of IODs in Figure 3, commensurate with the wide range of emissions of the various species considered. The maximum IOD modelled from emissions of CFC-11, which includes increases in the banks and subsequent release, is about 230 DU years (Figure 3; see also Table 1 for all the modelled IOD values) which, when divided by the integration length, corresponds to an average ozone depletion of about 3 DU, similar to the value seen in Figure 2. The largest average ozone depletion of around 15 DU is found for the case with the large CH3Br emissions (which has an IOD of 276 DU years over a perturbation of about 16 years duration). This value is again consistent with our results in Figure 2 where the short duration of the large average depletion arising from this short-lived halocarbon is evident.

To illustrate the advantage of IOD over the return date metric we look further at our integrations which study the impact of hypothetical illegal CFC-11 emissions. Nine different integrations explored increased CFC-11 (and possible CFC-12) emissions24. When an additional 35 Gg yr-1 of CFC-11 are emitted over 7 years (Scenario 1, Table 1), the IOD is about 29 DU years, with a global average loss of less than 1 DU and no discernible lengthening of the recovery period. The IOD, expressed as additional ozone loss above that calculated in the Base integration, is ~2%. So, while the delay in recovery is essentially zero, consistent with other model studies considering the recently observed CFC-11 emission increases24,25,26, the IOD does quantify the small increase in ozone depletion. In a second scenario with annual emissions more than twice the current values for 30 years (Scenario 9, Table 1), the IOD is 348 DU years, the global average loss is about 5 DU and the delay calculated in return date is about 10 years. In this scenario, both metrics (IOD or return date) do indicate the severity of the loss that would follow such emissions. Finally, turning to a scenario with 380 Gg yr-1 of CH3Br emissions over 5 years, we calculate an IOD of 233 DU years and an average loss of about 17 DU but there is no delay in the return date. In this case, the IOD is 16% of the Base, representing a severe impact, close to that of the extreme CFC-11 scenario, despite there being no delay in recovery showing that the return date metric fails completely to demonstrate the threat posed by such large CH3Br emissions, which is however captured starkly by IOD.

The IOD and ODP metrics both depend on estimating ozone depletion due to a specified emission of an ODS. Unlike ODP, however, IOD is an absolute measure of impact (like the delay in return date). ODP is a measure of a species’ impact on ozone, relative to CFC-11, and IOD, as a percentage of Base, is a measure of impact relative to all previous halogen emissions. This results in a clear relationship between the IOD and different methods for estimating ODPs (see Methods for further details) which can be used to compare emissions in a relative sense if required.

The empirical relationship in Equation 1 allows the IOD to be calculated simply for any halogenated species whose atmospheric and stratospheric lifetimes are known, given a choice of emission strength. For example, the unexpected emissions of CFC-11 during 2012-2019 are estimated to be between 120 and 440 Gg (Eeq of 0.092 and 0.34 Tg(Cl) respectively), which corresponds to an IOD of between approximately 9 and 34 DU years, small values which emphasise the huge importance of the early detection of the unexpected emissions of this long-lived gas. Emissions of DCM, a short-lived halocarbon that is not regulated under the Montreal Protocol, increased by approximately an annually-averaged 300 Gg yr-1 between 2007 and 201727. Given a tropospheric lifetime of 180 days1 and a stratospheric lifetime of 1-2 years12, the total lifetime is 142 days. In this case, we calculate an IOD for DCM of about 55 DU years (~4% of the Base IOD) associated with the increased emissions over this time period. This value for DCM depends sensitively on the lifetimes which are not well constrained; for example, simply using the quoted range for the stratospheric lifetime leads to changes in the IOD of about ±15%. Furthermore, we did not include any contribution from Cl-gases produced from the oxidation of DCM in the troposphere, which are subsequently transported to the stratosphere. These would increase the ozone depletion but for DCM are estimated to be less than 25% of the total reaching the stratosphere28. Nevertheless, despite the uncertainty, the magnitude of the estimated IOD supports concerns about future increases in DCM emissions.

The IOD depends on a number of variables – emissions and the release in the stratosphere, which depends on the whole atmosphere and stratospheric lifetimes. These latter factors represent probably the largest source of uncertainty for short-lived species since they depend *inter alia* on the location of the emissions, and on meteorological variability. The accumulation of reactive halogens in the stratosphere will also depend, as mentioned above, on the extent to which tropospheric degradation products of the halocarbons are themselves subsequently transported into the stratosphere29. For brominated species, the choice of efficiency factor will also be important and will vary with time, depending on other emissions21.

Calculation of the IOD using Equation 1 also depends, of course, on the value of *K* which must be evaluated using an atmospheric model so there will inevitably be inter-model differences in its value. This study has outlined the method for deriving the IOD. A model intercomparison exercise could be used to determine the most representative value for *K* and its uncertainty.

Although the IOD is specifically aimed at assessing the impact of emissions of halogenated species, the evaluation of IOD within a 3D model can also be used to quantify the impact of other perturbations to the stratosphere. For example, the IOD can be used to compare the impact on ozone layer recovery of different GHG emissions scenarios that are assessed within CCM simulations. It would be straightforward to evaluate the IOD from standard CCM output and would give a more meaningful measure of the impact of different GHG scenarios (e.g. for surface UV flux) than the simple delay to a return date.

We believe that the integrated ozone depletion metric in Figure 3 is a useful additional metric for both policy makers and scientists, being clearly superior in some circumstances to an assessment of the time delay in the return of ozone to a particular value. Furthermore, for gases whose lifetimes are known it provides a simple means of assessing the particular impact, and its importance to policy objectives, of any given emission scenario.

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| --- | --- | --- | --- | --- | --- |
| Simulation | Species emitted | Annual emission increment | Duration of emission | IOD(whole integration) | IOD(limited duration) |
| Base | Follows emissions of the WMO (2014) A1 scenario |
| Scenario 1 | CFC-11 | 35 Gg yr−1 | 7 years | 29.3 | 29.3 |
| Scenario 2 | CFC-11 | 35 Gg yr−1 | 15 years | 89.3 | 89.3 |
| Scenario 3 | CFC-11 | 35 Gg yr−1 | 30 years | 98.4 | 98.4 |
| Scenario 4 | CFC-11 | 15 Gg yr−1 + bank emission | 7 years | 34.4 | 34.4 |
| Scenario 5 | CFC-11 | 15 Gg yr−1 + bank emission | 15 years | 111.4 | 111.4 |
| Scenario 6 | CFC-11 | 15 Gg yr−1 + bank emission | 30 years | 231.7 | 231.7 |
| Scenario 7 | CFC-11 and CFC-12 | 15 Gg yr−1 + bank emission | 7 years | 97.6 | 97.6 |
| Scenario 8 | CFC-11 and CFC-12 | 15 Gg yr−1 + bank emission | 15 years | 186.0 | 186.0 |
| Scenario 9 | CFC-11 and CFC-12 | 15 Gg yr−1 + bank emission | 30 years | 348.4 | 348.4 |
| Scenario 10 | CH3CCl3 | 1000 Gg yr−1 | 1 year | -13.4 | -16.1 |
| Scenario 11 | CH3CCl3 | 1000 Gg yr−1 | 5 years | 84.9 | 66.4 |
| Scenario 12 | CH3CCl3 | 1000 Gg yr−1 | 10 years | 113.1 | 118.4 |
| Scenario 13 | CH3CCl3 | 5000 Gg yr−1 | 1 year | 65.9 | 45.0 |
| Scenario 14 | CH3Cl | 16000 Gg yr−1 | 1 year | -44.3 | -8.5 |
| Scenario 15 | CH3Cl | 16000 Gg yr−1 | 5 years | 30.4 | 54.1 |
| Scenario 16 | CH3Br | 380 Gg yr−1 | 1 year | 105.4 | 76.7 |
| Scenario 17 | CH3Br | 380 Gg yr−1 | 5 years | 310.9 | 275.9 |

Table 1: Summary of simulations used in this study. The Base simulation, run from 1960-2100, follows the experimental design of the WCRP/SPARC Chemistry Climate Model Initiative REF-C2 experiment and uses the RCP6.0 scenario for future greenhouse gas concentrations and the WMO (2014) A1 scenario for future ODSs concentrations. The Scenario simulations, run from 2012 to 2100 also follow these emissions scenarios, but include an additional halogen emission pulse. For each scenario the species, magnitude and duration of this emissions pulse is given in the table. For the Scenario simulations, all the additional halogen emissions start in 2012. IODs are in DU years.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Species emitted | $$τ\_{atmos}$$*(Modelled)* | $$τ\_{atmos}$$*(Literature)* | $$τ\_{strat}$$*(Modelled)* | $$τ\_{strat}$$*(Literature)* |
| CFC-11 | 55.7 | 52.0 | 55.7 | 55.0 |
| CFC-12 | 103.0 | 102.0 | 103.0 | 103.0 |
| CH3CCl3 | 4.4 | 5.0 | 41.6 | 38.0 |
| CH3Cl | 1.2 | 0.9 | 38.3 | 30.4 |
| CH3Br | 1.3 | 0.8 | 32.1 | 26.3 |

Table 2: Whole atmosphere (*τatmos*) and stratospheric (*τstrat*) lifetimes (years) of CFC-11, CFC-12, CH3CCl3, CH3Cl and CH3Br calculated using the UM-UKCA model and taken from the literature (WMO, 2018)1.

**Figures legends**



**Figure 1.** **A schematic of ozone recovery under different halogen emission scenarios based on chemistry-climate model calculations.** Global mean column ozone (DU) is plotted against time from 1960 to 2100. The black curve represents a Base case where the Montreal Protocol schedule is followed. The blue curve represents a case where some additional emissions of a long-lived ODS occur, while the red curve is representative of emission of a short-lived halocarbon. The blue and red shaded areas are the integrated ozone depletions (IODs) consequent on the additional emissions. The yellow shaded area is the IOD for the Base case relative to some historical (or projected future) value, integrated from the time that the additional emissions occur.



**Figure 2. Total column ozone trajectories calculated in our chemistry-climate model.24** The black lines show the Base case, assuming full compliance with the Montreal Protocol. Thin coloured lines show annual mean TCO from simulations with additional emissions of CFC-11 (green), CH3CCl3 (MCF; blue) and CH3Br (brown), where the legend gives the annual emission strength and the duration of the emissions, commencing in 2012. Thicker curves show the same data smoothed with an 11-point boxcar smoothing.



**Figure 3. Relationship between integrated ozone depletion (IOD), total halogen emissions and the stratospheric and whole atmosphere lifetimes.** Integrated ozone depletion (in DU years) from all UM-UKCA integrations is plotted against the total halogen emission in Tg Cl, EEq, multiplied by the whole atmosphere lifetime of the emitted species divided by its stratospheric lifetime. The correlation coefficient, r, is 0.84 and the grey shading represents the 95% confidence interval around the line of best fit. Even when the scenario with the largest emission of CFC-11 and CFC-12 is not included, the correlation coefficient remains high (0.75). The magnitude and duration of emissions is given in the legend. Closed circles represent scenarios in which emissions are assumed to go directly into the atmosphere. Open circles represent scenarios in which 75 Gg yr-1 goes into a bank and the remaining 15 Gg yr-1 is emitted directly. IOD uncertainty is estimated using an ensemble of UM-UKCA simulations following the same surface boundary conditions (i.e., emissions), which indicate that IOD differences can be as large as 48 DU years for simulations with the same halogen emissions over the time range considered here (2012-2100).

**Methods**

**Simulations**: In this study, we use simulations performed with the UM-UKCA chemistry-climate model (CCM). The UM-UKCA model consists of the HadGEM3-A configuration of version 7.3 of the Met Office's Unified Model30 coupled to the United Kingdom Chemistry and Aerosol scheme that explicitly considers individual halogen source gases31. The model has a horizontal resolution of 2.5° latitude × 3.75° longitude, and 60 vertical levels following a hybrid sigma-geometric height coordinate, extending from the surface to a model top at 84 km.

A baseline simulation (Base) was run from 1960 to 2100 following the experimental design of the WCRP/SPARC Chemistry-Climate Model Initiative (CCMI) REF-C2 experiment32, using the RCP6.0 scenario for future greenhouse gas concentrations and the WMO (2014) A1 scenario for future ODSs concentrations33. A further 17 scenarios were performed, running from 2012 to 2099, using a range of halogenated ODS lower boundary conditions. These scenarios considered emission of CFC-11, CFC-12, CH3CCl3, CH3Cl, and CH3Br in addition to that already assumed in the WMO (2014) A1 scenario. The emission strength and duration of these scenarios are provided in Table 1. Timeseries of global averaged total column ozone for each of the simulations is shown in Extended Data Figure 1.

**Calculating the stratospheric release**: The tropospheric and stratospheric lifetimes of a species, Χ, are related to its overall atmospheric lifetime by

$$\frac{1}{τ\_{atmos}}=\frac{1}{τ\_{strat}}+ \frac{1}{τ\_{trop}} $$

and the overall atmospheric loss rate of the species is given by

$$\frac{dX\_{a}}{dt}= \frac{dX\_{strat}}{dt}+ \frac{dX\_{trop}}{dt}$$

$$\frac{dX\_{a}}{dt}= - \frac{X\_{a}}{τ\_{strat}}-\frac{X\_{a}}{τ\_{trop}}= - \frac{X\_{a}}{τ\_{atmos}}$$

where Χa is the globally averaged abundance of Χ. So, the ratio of the amount of Χ destroyed in the stratosphere to the amount destroyed in the whole atmosphere is:

$$\frac{dX\_{strat}}{dX\_{a}}= \frac{τ\_{atmos}}{τ\_{strat}}$$

**Estimating uncertainty in IOD:** IOD is influenced not just by halogen emissions, but also by differences between model simulations arising from dynamical variability. We estimate the impact of this variability on our IOD calculations using an ensemble of UM-UKCA simulations performed with the same surface boundary conditions (REF-C2 simulations performed as part of the CCMI project32). As these simulations have the same surface boundary conditions, stratospheric halogen changes do not explain the IOD differences between these simulations. It was found that these scenarios could differ by up to 48 DU years over the time range considered here (2012-2100), and we use this value in Figure 3 as an estimate of the uncertainty in IOD.

We have calculated the IOD over the length of our integrations (see Figure 3). However, once several lifetimes of the perturbation have passed, any differences between the calculated IOD and the IOD from the Base integration will arise purely from the variability inherent in our chemistry/climate integrations. We did find, in the case of the short-lived gases included in our UM-UKCA integrations, that differences from Base were calculated long after the halogen perturbation had decayed. For this reason, the IOD could also be calculated just over the perturbation period, which for short-lived gases we took to be the duration of the emission pulse plus 5 times the whole atmospheric lifetime (τatmos) plus the time for removal from the stratosphere (for which we used 7 years, the typical maximum mean age of air in the polar lower stratosphere22,23). So, for a scenario in which CH3Cl (modelled τatmos = 1.2 years) is emitted for 5 years, the IOD would be calculated over a time horizon of 18 years. Extended Data Figure 2 plots IODs calculated in this way (with the IODs for long-lived gases calculated as previously) versus the total halogen emission in Tg Cl, EEq, multiplied by τatmos/τstrat. In this case the correlation coefficient is improved, relative to Figure 3, to r=0.90, but the gradient remains the same (*K* = 100±11 DU/Tg(Cl)).

**Relationship to Ozone Depletion Potential (ODP)**: The ODP is a measure of the stratospheric ozone destroyed per unit mass of emissions of a long-lived source gas compared to CFC-11. It can be estimated directly from models for any emission scenario or, in the case of continuous steady-state emissions, by the semi-empirical ODP. In the case of model-based estimates, there is clearly a correspondence between the IOD and ODP. They are both based on the calculation of ozone depletion in the model following an assumed emission of the ODS (and in the case of ODP, a similar emission of CFC-11), although as an absolute measure of impact of emissions on ozone, the IOD is advantageous for application to ozone recovery. For ODP estimates which are partly based on empirical quantities, we can demonstrate the correspondence between IOD and ODP. For example, the semi-empirical ODP is given by29

$$ODP\_{i}=φ\_{i} α\_{i} \frac{τ\_{i}}{τ\_{cfc11}}\frac{M\_{cfc11}}{M\_{i}}\frac{N\_{i}}{3}=φ\_{i} α\_{i} \left(\frac{τ N}{M}\right)\_{i}/\left(\frac{3 τ\_{cfc11}}{M\_{cfc11}}\right)$$

where *φ* is the mean fractional release in stratosphere of species *i* compared to CFC-11, *α* is the relative efficiency factor for bromine species (= 1 for chlorine species), *τ* is here the total atmospheric lifetime, *M* is the relative molecular mass and *N* is the number of Cl (or Br) atoms in the molecule. Our relationship (Equation 1) for the IOD of a species *i* derived from the CCM results is:

$$IOD=K E\_{Eq} \left(\frac{τ\_{atmos}}{τ\_{strat}}\right) $$

where *K* is the slope of Figure 3, *τatmos* and *τstrat* are the atmospheric and stratospheric lifetimes, respectively, and *EEq* is the total equivalent Cl emissions. As different species have the same slope in Figure 3 we can compare the ratio of integrated ozone depletion due to emissions of species *i* and CFC-11 as:

$$\frac{IOD\_{i}}{IOD\_{CFC11}}= \frac{τ\_{atmos,i} }{τ\_{strat,i} } \frac{E\_{Eq,i}}{E\_{Eq,CFC11}}= α\_{i} \frac{M\_{CFC11}}{M\_{i}} \frac{N\_{i}}{3} \frac{τ\_{atmos,i}}{τ\_{strat,i}} \frac{E\_{i}}{E\_{CFC11}} $$

where *Ei* is the emitted mass of species *i* and given that for CFC-11 *N* = 3, *α* = 1 and, as there is no tropospheric loss, *τatmos* = *τstrat*. This ratio is related to the semi-empirical ODP for species *i* by:

$$ \frac{IOD\_{i}}{IOD\_{CFC11}}=ODP\_{i} \frac{E\_{i}}{E\_{CFC11}} \frac{τ\_{CFC11}}{τ\_{strat,i}} \frac{1}{φ\_{i}} $$

For equal mass emissions of a species with no tropospheric loss (*τatmos* = *τstrat*) and similar stratospheric destruction rate to CFC-11 (*ϕ* = 1), there is a simple correspondence between the ratio of the IODs and the ODP. Note, however that the IOD provides an absolute measure of the ozone impact and can incorporate any time-varying emission scenario. Moreover, the IOD implicitly deals with cases such as VSLS where the semi-empirical ODP approach is not valid.

The above analysis relates IOD to the semi-empirical ODP in the case of steady state emissions; other definitions of ODP exist which could also be related to IOD29.

**Data availability**

The simulation data used in this study are archived within the UKCA group workspace on the JASMIN platform ([www.jasmin.ac.uk](http://www.jasmin.ac.uk)) maintained by the Centre for Environmental Data Analysis (CEDA) and are available on reasonable request from the corresponding authors.

**Code availability**

The UM-UKCA model is available for use under licence. A number of research organisations and national meteorological services use the UM in collaboration with the Met Office to undertake basic atmospheric process research, produce forecasts, develop the UM code, and build and evaluate Earth system models. For further information on how to apply for a licence see <http://www.metoffice.gov.uk/research/modelling-systems/unified-model> (last access 27 September 2021).

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**Author contributions** JAP proposed the initial study which was developed jointly by JK, JAP and MPC. JK performed most of the calculations with support from NLA and PTG. JAP and JK led the writing to which all authors contributed.

**Competing interests** The authors declare no competing interests.

**Additional Information**

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Extended Data Figure Legends



**Extended Data Figure 1: Time series, from 1960 to 2100, of total column ozone (TCO, DU), averaged from 90°S-90°N, from the simulations analysed in this study.** Part a) plots annual mean model data and b) plots the data after it has been smoothed using an 11-point boxcar smoothing, following Dhomse et al34. The large difference at 2100 between Base and the scenario with 30-year emissions of CFC-11 and CFC-12 arises from the large increase in (and subsequent emission from) their banks, assumed in that scenario (as discussed in Keeble et al.24).



**Extended Data Figure 2: Integrated ozone depletion (IOD, in DU years) from all UM-UKCA integrations plotted against the total halogen emission in Tg Cl, EEq, multiplied by the whole atmosphere lifetime of the emitted species divided by its stratospheric lifetime, for comparison with Figure 3.** The IOD for short-lived gases is calculated over the perturbation period as defined in the text, otherwise as in Figure 3. The correlation coefficient, r, is now 0.90, and when the scenario with the emission of CFC-11 and CFC-12 is not included, the correlation coefficient remains high (0.83).