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Key Points:

- Between 1955 and 2005, global warming from ozone-depleting substances (ODSs) was more than half of that due to CO_2, and about one third of total anthropogenic warming
- ODSs have been about 20% more effective than CO_2 at causing global warming per unit radiative forcing
- ODS warming peaks in the Arctic and is 66% of that due to CO_2, while their Effective Radiative Forcing (ERF) opposes Arctic amplification more than the CO_2 ERF

Large Contribution of Ozone-Depleting Substances to Global and Arctic Warming in the Late 20th Century

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Abstract While previous studies have suggested a substantial role of ozone-depleting substances (ODSs) in historical climate change, their relative contribution to historical anthropogenic warming has not been quantified before. Analyzing all-but-one-forcing, 20-member ensembles of historical simulations with a state-of-the-art Earth System Model, we find that over the 1955–2005 period ODSs are responsible for 30% of global warming, 37% of Arctic warming, and 33% of summertime Arctic sea ice loss. Effective Radiative Forcing (ERF) calculations reveal that the global warming response to ODSs per unit of ERF is about 20% larger than for CO_2, which may be due to stronger feedbacks and the difference in temporal evolution with ODSs having leveled off and CO_2 still increasing in 2005. While the response to both peaks in the Arctic, the ODS ERF opposes Arctic amplification more than the CO_2 ERF. Our findings highlight the importance of the Montreal Protocol for mitigating future climate change.

Plain Language Summary Ozone-depleting substances (ODSs) are chemicals developed in the 1920s and 1930s for use in spray cans, refrigerators and plastic foams. Their commercial use increased rapidly in the 1950s and 1960s, but their phase out is underway since the signing of the Montreal Protocol in the 1987, following the identification of their devastating impact on the stratospheric ozone layer. It is well known that ODSs are powerful greenhouse gases, with the second largest warming effect between 1955 and 2005. However, their relative contribution to past global warming has not been quantified previously using comprehensive climate models. Here we show that ODSs were responsible for roughly a third of late 20th Century global warming, Arctic warming and Arctic sea ice decline. In addition, we find that the impact of ODSs on global temperatures is about 20% larger than expected based on the impacts they have on the radiative balance. The impacts of ODSs peak in the Arctic, while their radiative forcing peaks in the tropics, and thus opposes Arctic warming amplification. These findings enhance our understanding of drivers of past climate change, and highlight the importance of the Montreal Protocol for future climate change mitigation.

1. Introduction

Ozone-depleting substances (ODSs), which include chlorofluorocarbons (CFCs), are organic halogen compounds known to be the main cause behind the formation of the stratospheric "ozone hole" in the late 20th century. Such compounds were developed in the 1920s and 1930s for use in aerosol propellants, plastic foams, and refrigerants. The atmospheric concentrations of ODSs started to increase rapidly in the 1950s and 1960s, and their phase out was internationally agreed upon with the signing of the 1987 Montreal Protocol, only a few years after the discovery of their devastating impact on the ozone layer (Farman et al., 1985). While ODSs are best known for their chemical impacts on ozone, they are also known to be potent greenhouse gases (GHGs) (Ramanathan, 1975). In fact the phasing out of ODSs under the Montreal Protocol will result in a substantial mitigation of climate change (Velders et al., 2007): the avoided global warming has been estimated to reach 1°C by 2050 (Goyal et al., 2019), a considerable amount in the context of the 2015 Paris Agreement, which aims to keep global mean warming to well below 2°C above pre-industrial levels.

But here we ask: how much of ODS-driven global warming has *not* been avoided? In other words: how much actual warming have ODSs caused prior to their stabilization and eventual phase out? This question has not received much attention, as ODSs are considered to be minor GHGs, based on their relatively small radiative
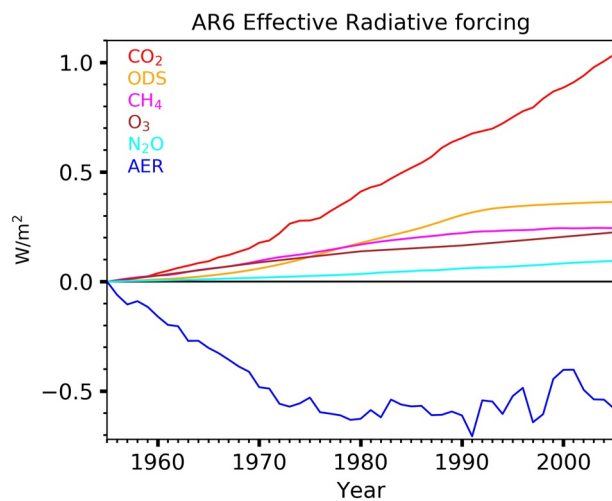
Figure 1. The effective radiative forcing relative to 1955 due to CO_2, ODSs, CH_4, total ozone (O_3, which is mainly due to tropospheric ozone), N_2O, and anthropogenic aerosols (AER), as assessed by the Intergovernmental Panel on Climate Change Sixth Assessment Report (P. Forster et al., 2021; Smith et al., 2021).

forcing (RF) when computed relative to pre-industrial levels. However, it was already pointed out in the first assessment report of the Intergovernmental Panel on Climate Change (IPCC) that RF associated with CFCs was responsible for 24% of the change in RF between 1980 and 1990. It was recently brought to light that, over the period of their largest increase (between 1955 and 2005), ODSs collectively are the second largest GHG forcing agent after carbon dioxide (Polvani et al., 2020). Figure 1 shows the currently best available estimates of Effective Radiative Forcing (ERF) of all major climate forcers, as assessed by the IPCC Sixth Assessment Report (AR6) (P. Forster et al., 2021; Smith et al., 2021). Over the 1955–2005 period, the ERF of ODSs exceeds the ERF of all non-CO_2 GHGs, notably methane, nitrous oxide, and tropospheric ozone.

Climate impacts of this large ODS RF were recently quantified by Polvani et al. (2020) in two versions of the climate model developed at the National Center for Atmospheric Research. They reported that for historical simulations in which ODSs are fixed to 1955 levels, the global mean 1955–2005 surface warming is 30% smaller—and the Arctic mean surface warming and September sea ice loss are 50% smaller—than in standard historical simulations in which all forcings (including ODSs) follow the observed time evolution. That study suggested a potentially important role for ODSs in historical warming, but its conclusions remain to be confirmed with an independent model. Furthermore, Polvani et al. (2020) were unable to quantify the relative contribution of ODSs to historical anthropogenic warming. This is because the net warming results from a cancellation between anthropogenic warming associated with GHGs and ozone, and anthropogenic cooling associated with aerosols. To properly quantify the contribution of ODSs to historical anthropogenic warming requires simulations that were not available at the time. The first goal of this study, therefore, is to validate the findings of Polvani et al. (2020) using a different model, and extend them by quantifying the relative contribution of ODSs to late 20th Century anthropogenic warming.

Furthermore, in trying to explain the surprisingly large warming from ODSs, one is led to ask whether ODSs might be more efficient at warming the global climate than other forcing agents. This question is typically addressed by calculating the "efficacy" E of a forcing agent (Hansen et al., 2005), here defined as the global temperature response ΔT per unit RF F of that agent, relative to carbon dioxide, that is,

$$E = \frac{\Delta T / F}{\Delta T_{CO_2} / F_{CO_2}}. \tag{1}$$

In this expression the RF F is specified as the ERF over a period of interest, that is, the radiative energy imbalance after taking into account rapid adjustments, as recommended by P. M. Forster et al. (2016).

The efficacy of nearly all forcing agents, including ODSs, has recently been estimated in a recent multi-model study. Richardson et al. (2019) have reported a value of E close to unity for CFC-11 and CFC-12 (and, in fact, for all GHGs). However, it is important to realize that this result was obtained from simulations with an unrealistically large perturbation from present-day concentrations of ODSs; in order to produce a strong forcing signal against large internal variability, concentrations of ODSs at year 2000 levels were multiplied by a factor of between 4 and 9 in that study. This is highly unrealistic, as ODS concentrations have peaked around the year 2000, and have been decreasing since as a result of the Montreal Protocol. In addition, and perhaps more importantly, Richardson et al. (2019) focused on the equilibrium response to an instantaneous forcing, whereas the climate system is far from equilibrated at this time. Therefore, it is unclear whether such equilibrium calculations are in fact quantitatively applicable to the highly transient historical climate change. Hence, the second goal of this study is to quantify the efficacy of ODSs within a realistic, transient evolution of the climate system. In contrast to Richardson et al. (2019), we here bring out the signal from the noise not by imposing large and unrealistic levels of ODSs, but using a large ensemble of realistic transient runs with time dependent ODS evolution as in the observations.

2. Model and Simulations

The model employed in this study is the Canadian Earth System Model version 5 (CanESM5) (Swart et al., 2019), a state-of-the-art Earth System Model that has extensively contributed to CMIP6, the Phase 6 of the Coupled Model Intercomparison Project (Eyring et al., 2016). While CanESM5 is one of the models that has been reported to feature a higher climate sensitivity than its counterpart from the Coupled Model Intercomparison Project Phase 5 (CMIP5) (Virgin et al., 2021; Zelinka et al., 2020), we note that over the time period that this study focuses on (1955–2005), simulated climate trends are consistent with observations (see Results).

Several 20-member ensembles of CanESM5 simulations are considered in this study. First, we analyze an ensemble of 20 simulations with standard CMIP6 historical forcings from 1850 to 2014, each member of which is run with identical forcings but with different initial conditions obtained from the preindustrial simulation at 50-year intervals. Second, for this study we have performed four additional 20-member ensembles, all branched from the year 1955 of the corresponding historical simulations, in which we separately keep (a) ODSs, (b) CO_2, (c) anthropogenic aerosols, and (d) ODSs plus stratospheric ozone, fixed at 1955 values. The "response" to a forcing relative to 1955 is then computed as the ensemble-mean difference between simulations in which that forcing agent is held fixed at 1955 values and the corresponding standard historical simulations (in which that agent follows the observed evolution). For each forcing, any "change" from 1955 to 2005 presented in this letter is computed as the difference between the 2001–2005 mean in the simulations in which the forcing is fixed and the historical simulation.

An important novel aspect of this study is that we place the climate warming from ODSs in the context of the total anthropogenic warming (labeled "AntW"), which is defined here as changes due to all anthropogenic forcings minus the cooling from anthropogenic aerosols. This total anthropogenic warming hence includes the impacts of all GHGs as well as ozone. It is quantified here by the 1955–2005 changes in the simulations with fixed aerosols (assuming that the impact of natural forcings over the period 1955–2005 is negligible).

To calculate the time evolution of the ERF associated with each forcing agent, we have performed complementary 20-member ensembles of atmosphere-only simulations (except for the case in which both ODSs and stratospheric O_3 are fixed to 1955 levels), spanning the 1955 to 2005 period. For this, we first calculated the ensemble mean monthly varying climatologies of sea-surface temperature and sea ice concentration, averaged over the 1950–1960 period from the historical coupled simulations, and then prescribed these to all atmosphere-only simulations. The ERF associated with a forcing agent relative to 1955 is then computed as the difference in the top-of-atmosphere net radiative flux between the simulation in which that agent is held fixed to 1955 values and the standard atmosphere-only historical simulation (in which that agent follows the observed evolution).

Lastly, to validate our model results, the CanESM5 ERFs thus computed are compared to the currently best available estimates of the ERF as assessed by the AR6, the IPCC Sixth Assessment Report (P. Forster et al., 2021; Smith et al., 2021). AR6 ERFs were calculated from modeled and observational data. More details on the CanESM5 radiation scheme (von Salzen et al., 2013), used forcings and AR6 ERFs can be found in the Supplementary Information.

3. Results

We start by highlighting the fact that the 1955–2005 global mean surface air temperature (GSAT) changes in the historical CanESM5 simulations are consistent with observed changes, as the CanESM5 ensemble range (gray error bars, Figure 2a) encompasses the observed warming. In addition, we find that the corresponding ERF computed from the atmosphere-only simulations are consistent with the current best estimates from the IPCC AR6, both for the response to all forcings and the response to CO_2 and aerosols separately (Figure 2b). The only exception is that the ERF associated with ODSs in CanESM5 (0.48 W/m^2) is slightly higher than that estimated from AR6 (0.36 W/m^2).

We next consider the 20-member ensemble of simulations in which ODSs are fixed to 1955 levels. We find that 1955–2005 global warming in those simulations is 41% (36%–45%, 5%–95% confidence range) smaller than in the standard historical simulations (in which ODSs follow the observed evolution). This reduction is slightly larger than but similar to the 34% value reported in Polvani et al. (2020)—who used a different comprehensive climate model, slightly different historical forcings (CMIP5 instead of CMIP6), and a smaller ensemble size (10
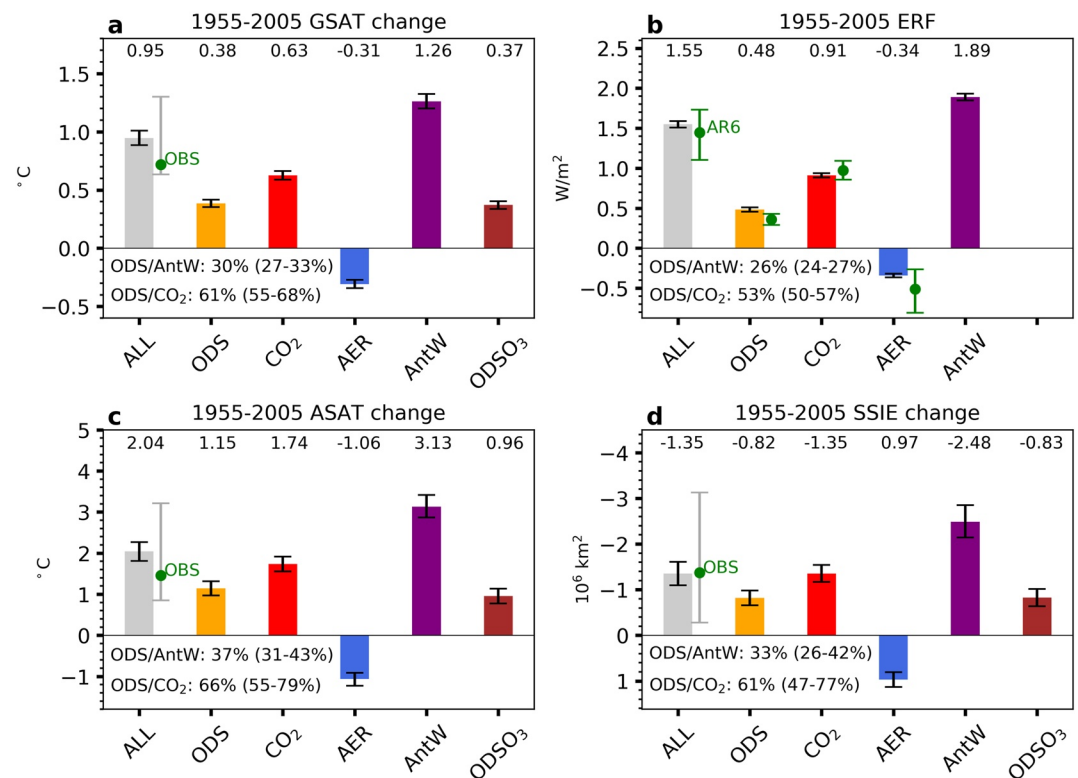
Figure 2. The Canadian Earth System Model version 5 (a) global mean surface air temperature (GSAT) change, (b) effective radiative forcing (ERF), (c) Arctic mean surface air temperature (ASAT) and (d) September Sea Ice Extent (SSIE) change over the 1955–2005 period associated with all historical forcings (ALL) and ODSs, CO_2, aerosols (AER), anthropogenic warming agents (AntW) and ODSs plus stratospheric ozone ($ODSO_3$) separately. The black error bars represent the 5%–95% confidence range of the ensemble mean as determined by bootstrapping, the gray error bars in (a, c, d) the ensemble range and the numbers on top the ensemble mean values. The green dots in (a, c) denote the observed values from HadCRUT5 (Morice et al., 2021), the green dot in (d) denotes the observed values from Walsh et al. (2015), and the green dot and lines in (b) represent the best estimate and 5%–95% confidence range as assessed by the Intergovernmental Panel on Climate Change Sixth Assessment Report (P. Forster et al., 2021; Smith et al., 2021).

instead of 20)—suggesting that this result is not very sensitive to model formulation or details of the forcing. The difference between the fixed and varying ODS simulations is displayed in orange in Figure 2a and shows that in the ensemble mean, 0.38°C of the warming over the 1955 to 2005 period can be attributed to ODSs.

But how does this warming from ODSs compare with the warming from CO_2 over the same period? To answer this, we analyze the 20-member ensemble with CO_2 fixed at 1955 level. We find that the warming from CO_2 is 0.63°C. This implies that the warming from ODS is *more than half* (61%, uncertainty range provided in Figure 2) of the warming from CO_2, the dominant GHG. This surprisingly large number constitutes the first important finding of our study.

Furthermore, we place the warming from ODSs in the broader context of the total anthropogenic warming (which includes well mixed GHGs and ozone, and excludes the cooling effects of aerosols, see the previous section). The warming from all anthropogenic forcings (labeled "AntW" in Figure 2) is found to be 1.26°C in the ensemble mean. ODSs, therefore, have contributed nearly one third (30%) of the total anthropogenic warming over the 1955 to 2005 period.

One may wonder if the large surface warming from ODSs is canceled, to some degree, by the depletion of stratospheric ozone that they induce. In terms of RF, a large cancellation was suggested in an influential early study (Ramaswamy et al., 1992), and more recently by Morgenstern et al. (2020); other studies, however have indicated that the cancellation is actually much smaller (see Chiodo and Polvani (2022) and discussion therein). To quantify the potential global warming cancellation from ozone depletion, we examine an additional 20-member ensemble in which *both* ODSs and stratospheric ozone are fixed at 1955 values. As seen in the last item in Figure 2a

(labeled "ODSO$_3$"), this additional ensemble reveals that the indirect impact of ODSs on GSAT via stratospheric ozone depletion is negligible, consistent with the relatively small RF of stratospheric ozone, and confirming a previous study (Polvani et al., 2020).

Given the large warming from ODSs, we now turn to the question of efficacy, that is, whether ODSs are more efficient than CO$_2$ at warming global temperatures per unit of RF. Time varying ERFs associated with individual forcing agents, calculated from fixed SST and sea-ice simulations, are plotted in Figure 2b. We find that while the ODS contribution to ERF is about a quarter of that from all anthropogenic warming agents, the ODS contribution relative to that of CO$_2$ is 53%, which is smaller than the ODSs to CO$_2$ ratio in GSAT change. In other words, our results show that the GSAT response to ODSs relative to that of CO$_2$ is larger than that expected from their relative ERFs. The ensemble mean ODS efficacy (Equation 1) is 1.19 (with a 5%–95% uncertainty range of 1.07–1.32), which, according to a one-sided t-test is statistically larger than unity ($p = 0.014$). We also note that about one third of the ensemble members show an ODS efficacy that is smaller than unity: this indicates the presence of large internal variability, and the need to perform large ensembles of simulations to establish a statistically significant efficacy value.

This second key result of our study, the high efficacy of ODSs, stands in contrast to the result obtained from highly idealized equilibrium forcing experiments (Richardson et al., 2019), which have reported an efficacy for CFC11 and CFC12 close to unity. Analyzing the realistic transient evolution of historical forcings over the 1955–2005 period, our model shows that ODSs are almost 20% more effective at warming global temperatures than carbon dioxide. Possible explanations for this result are offered in the Discussion section.

Next we turn to the spatial patterns and the time evolution of the surface temperature response caused by different forcings, shown in Figure 3. Comparison of the first two rows reveals that CanESM5 is able to reproduce the general features in the observations, with the largest warming over land in the Arctic, and a slight cooling prior to the 1970s followed by a rapid warming thereafter. The third and fourth rows in Figure 3 reveal that while the ODS impacts are slightly smaller than those due to CO$_2$, their spatial and temporal impacts are very similar, peaking in the Arctic with relatively little warming before the 1970s and rapid warming thereafter. Aerosol cooling (Figure 3, last row) also peaks in the Arctic, but shows a different time evolution, rapidly increasing between the 1950s and 1980s, followed by relatively small changes thereafter, which is consistent with previous studies (Fyfe et al., 2013). Together the CO$_2$, ODS and aerosol patterns explain most of the features in the historical simulations and observations, with the slight cooling prior to the 1980s attributable to anthropogenic aerosols, and the rapid warming that maximizes in the Arctic attributable to GHG increases. Comparison of the two rows in Figure S1 in Supporting Information S1 shows the indirect impacts of ODSs on surface temperature via ozone depletion are very small. Recall, however, that stratospheric ozone depletion has had large climate impacts on Southern Hemisphere climate (Previdi & Polvani, 2014), although these impact are not a consequence of direct RF, but of changes in the atmospheric circulation (e.g., the poleward migration of the midlatitude jet). Unlike stratospheric ozone, ODSs affect the climate system primarily via RF and surface warming. A careful comparison of the distinct impacts of ODSs and stratospheric ozone depletion on Southern Hemisphere climate will be reported in an upcoming paper.

Finally, since the ODS contribution to historical warming peaks in the Arctic, we take a closer look at the implications for Arctic temperature and sea ice. As seen in Figures 2c and 2d, respectively, the 1955–2005 Arctic warming and September sea ice decline in the historical CanESM5 simulations are consistent with observed changes, as the CanESM5 ensemble range encompasses the observed values. Fixing ODSs to 1955 levels reduces 1955–2005 Arctic warming by 55% (45%–68%) and September sea ice extent decline by 45% (18%–67%), in good agreement with the values reported in Polvani et al. (2020). ODSs are found to be responsible for 1.15°C of the Arctic mean warming, which amounts to 66% of Arctic warming due to CO$_2$, and to 37% of the Arctic warming that is due to all anthropogenic warming agents. The ratio of Arctic relative to the global mean warming, referred to as Arctic amplification factor, is 2.99 (2.49–3.53) for ODSs, which is slightly larger than for CO$_2$ (2.78, 2.45–3.12), consistent with a previous study (Liang et al., 2022). However the difference is not statistically significant in our model. As for September sea ice extent, we find that ODSs are responsible for 0.82 million km^2 of its decline, which is 33% of the decline due to all anthropogenic warming. In summary, more than a third of changes in key Arctic climate indicators between 1955 and 2005 can be attributed to ODS increases.

4. Discussion

One of the key results from this study is that over the period over which ODSs increased (between 1955 and 2005), the GSAT response to ODSs per unit of ERF is about 20% larger than the corresponding ratio for CO$_2$.
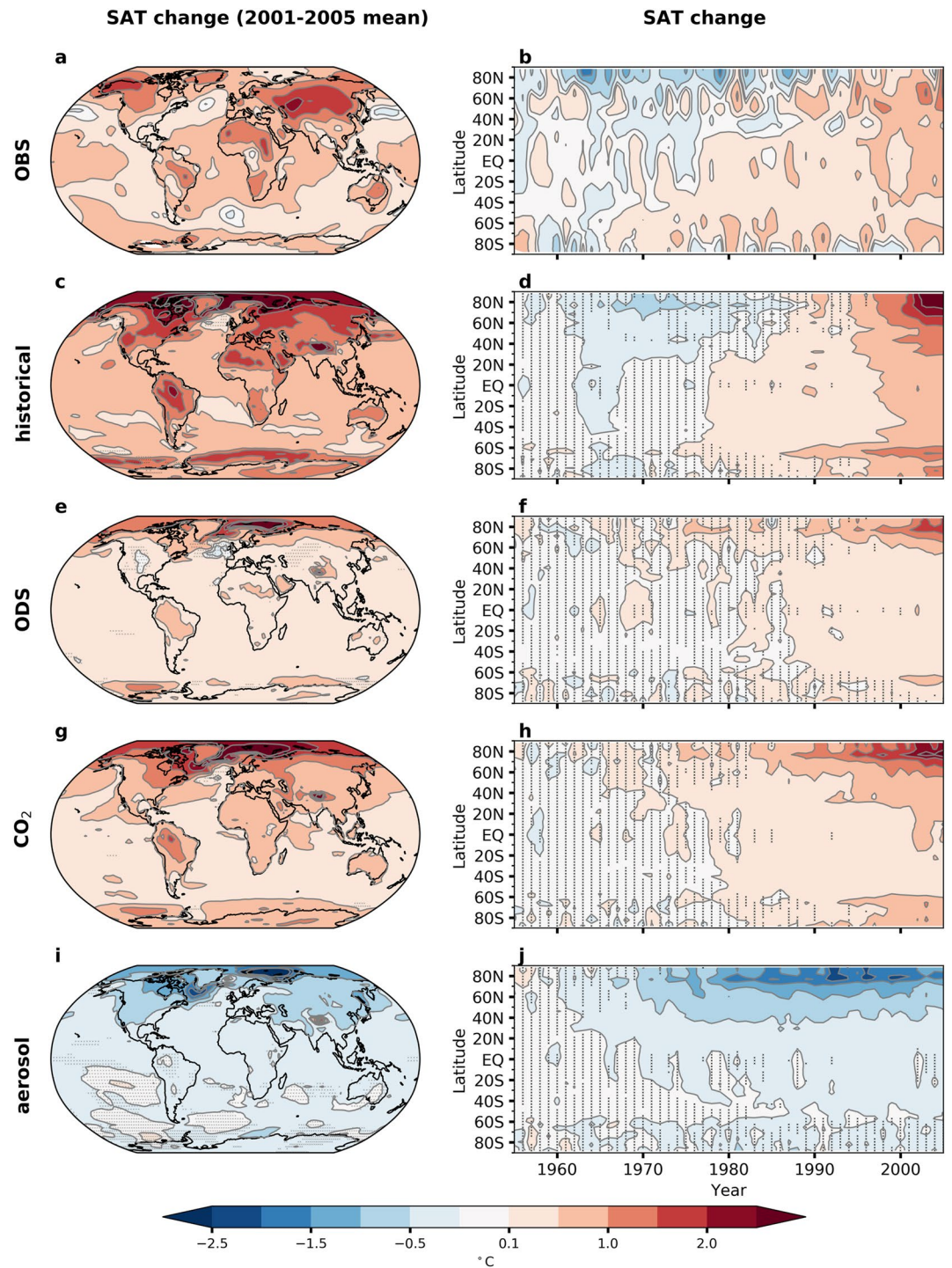
Figure 3. (left) The surface air temperature (SAT) change between the 1950–1960 and 2001–2005 averages and (right) the zonal mean SAT anomaly relative to 1955 in (a, b) observations, and the modeled response to (c, d) all historical forcings, (e, f) ozone-depleting substances, (g, h) CO_2 and (i, j) aerosol changes. Crosses in (c–j) indicate grid points with changes that are not statistical significant at the 95% level.

While a detailed analysis is beyond the scope of this study, we here provide two possible explanations. First, consider a very simple energy balance model (Boer & Yu, 2003; Gregory et al., 2004):

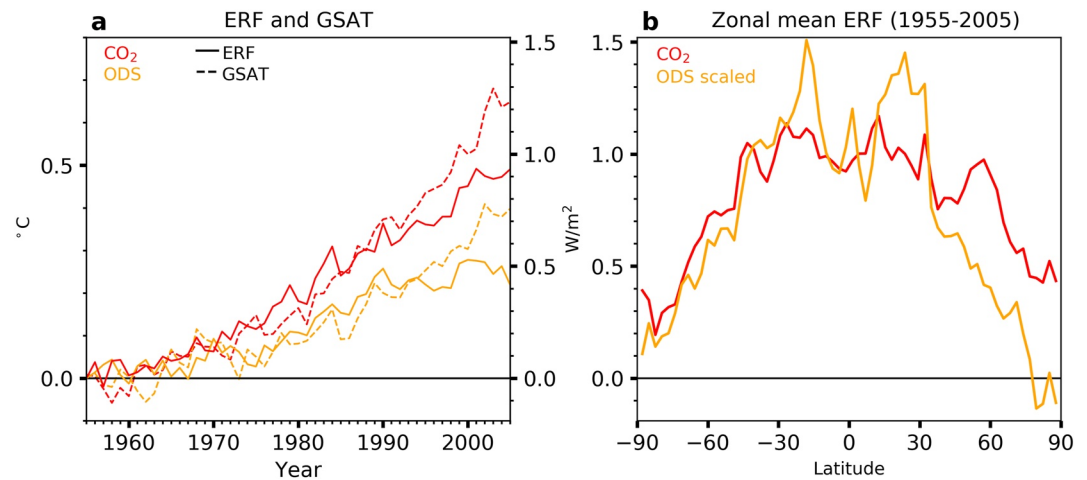$$N = F - \alpha \Delta T, \qquad (2)$$

Figure 4. (a) Timeseries of (solid) the effective radiative forcing (ERF) and (dashed) the global mean surface air temperature change relative to 1955 due to CO_2 and ozone-depleting substances (ODSs), and (b) the 1955–2005 zonal mean CO_2 ERF, and the zonal mean ODS ERF multiplied by the ratio of the global mean CO_2 and ODS ERFs.

where N is the top-of-atmosphere energy imbalance, ΔT is the GSAT response to the global mean forcing F, and α is the climate feedback parameter (larger values of α^{-1} correspond to stronger feedbacks). With CanESM5, for the 1955–2005 period, we find that α^{-1} associated with ODS increases is about 11% larger than that associated with CO_2 increases (1.43°C per W/m² for ODSs vs. 1.29°C per W/m² for CO_2). This finding indicates that ODSs are associated with inherently stronger feedbacks than CO_2, as suggested by Polvani et al. (2020). However, stronger feedbacks only explain roughly half of the 20% stronger GSAT response to ODSs per unit of ERF compared to that of CO_2.

We thus here propose a second factor that contributes to the enhanced ODS efficacy: the different *time evolution* of ODSs and CO_2 over the 1955 to 2005 period. The solid orange line in Figure 4a shows that after 1990 the ERF due to ODSs levels off (see Figure 1), as one expects from the time evolution of the ODS concentrations. But, despite the stabilization of the ODS ERF, the associated GSAT response continues to increase after 1990 (dashed orange line), as GSAT continues to adjust. The fact that surface temperature continues to rise even as the ERF from ODS stops rising yields in a stronger warming response per unit forcing as time progresses. In contrast, the CO_2 forcing continues to increase approximately linearly beyond 1990, and the GSAT response is not allowed to fully adjust to the higher levels of CO_2. This results in a smaller GSAT response to CO_2 per unit of ERF increase than to ODSs. In summary, we suggest that the different time evolution of the ODS and CO_2 ERF, and the associated GSAT response explains (at least part of) the enhanced ODS efficacy. This suggestion is supported by the fact that over the shorter 1955 to 1990 period, the period over which both ODSs and CO_2 both increase rapidly, the ODS efficiency at warming GSAT is very similar to that of CO_2.

The other key result of our study is that ODSs have contributed considerably to Arctic warming and sea ice loss in the second half of the 20th. This is particularly surprising since their RF opposes Arctic amplification, as shown in Chiodo and Polvani (2022). The RF calculations in Chiodo and Polvani (2022), however, did not take into account the rapid adjustments that are included in our ERF calculations. In Figure 4b, we show the zonal mean ERFs associated with ODSs and CO_2: these curves are qualitatively similar to those for RF in Chiodo and Polvani (2022), see their Figure 4b. The similarity between ERF and RF indicates that rapid adjustments are relatively unimportant for ODSs, as reported by Hodnebrog et al. (2020). Note that the ERF of ODSs opposes Arctic amplification even more than the ERF of CO_2, making their large contribution to Arctic climate change even more unexpected. This may be due to stronger local feedbacks as suggested by Liang et al. (2022).

In conclusion, using a comprehensive state-of-the-art earth system model, we have for the first time quantified the relative contribution of ODSs to historical anthropogenic warming, and found that roughly one third of the surface warming between 1955 and 2005 is attributable to ODSs. While the surface temperature changes attributable to ODSs in CanESM5 were found to be of similar magnitude as those reported in an earlier study with a different model (Polvani et al., 2020), the robustness of these results will need to be corroborated with

other models, especially in light of our finding that the CanESM5 ERF associated with ODSs may be slightly overestimated. Nevertheless, our results add to the mounting evidence that ODSs have played a substantial role in historical climate change, and highlight the fact that by phasing out the production and consumption of ODSs under the Montreal Protocol has not only halted the formation the Antarctic ozone hole, but has also played a crucial role in mitigating past and future climate change.

Data Availability Statement

Python scripts to create the figures are available at https://gitlab.com/michael.sigmond/cfc. The data used in this study is available at https://zenodo.org/record/6908225#.YuASoj3MI2x.

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