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Distinguishing the impacts of ozone-depleting substances and well-mixed greenhouse gases on Arctic stratospheric ozone and temperature trends

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Abstract Whether stratospheric cooling due to increases in well-mixed greenhouse gases (WMGHG) could increase the depletion of Arctic stratospheric ozone has been the subject of scientific and public attention for decades. Here we provide evidence that changes in the concentrations of ozone-depleting substances (ODS), not WMGHG, have been the primary driver of observed Arctic lower stratospheric trends in both ozone and temperature. We do so by analyzing polar cap ozone and temperature trends in reanalysis data: these clearly suggest that both trends are mainly driven by ODS in the lower stratosphere. This observation-based finding is supported by results from a stratosphere-resolving chemistry-climate model driven with time-varying ODS and WMGHG, specified in isolation and in combination. Taken together, these results provide strong evidence that ODS are the main driver of changes in the Arctic lower stratospheric temperatures and ozone, whereas WMGHG are the primary driver of changes in the upper stratosphere.

1. Introduction

Since the detection of the Antarctic ozone hole in the late 1980s [e.g., Farman et al., 1985], the state of the Earth’s stratosphere in polar regions has received much attention. It is now well established that man-made ozone-depleting substances (ODS) are the primary cause of stratospheric ozone depletion [e.g., Molina and Rowland, 1974; Rowland and Molina, 1975]. Societal awareness of this issue led to the successful implementation of the Montreal Protocol (and several subsequent amendments), which in turn led to the cessation of anthropogenic emissions of ODS (and thus ended increases in their atmospheric burdens): this has had a measurable effect on column ozone [e.g., Mäder et al., 2010; World Meteorological Organization (WMO), 2011]. For the future, current state-of-the-art chemistry-climate models consistently project a recovery (or even superrecovery) of column ozone to 1980 levels (or above) in the second half of the 21st century [Stratospheric Processes and their Role in Climate Chemistry-Climate Model Validation Activity (SPARC-CCMVal), 2010].

Several studies have demonstrated the crucial role of surface chemistry, on and in polar stratospheric clouds (PSCs), for polar ozone depletion [Peter, 1997; Solomon, 1999], especially in the formation of the ozone hole over Antarctica. Because these clouds form at particularly cold temperatures, the volume of air colder than the threshold for PSC formation (hereinafter $V_{PSC}$) is widely used as a convenient proxy for polar ozone loss. A number of studies have reported an upward trend in Arctic $V_{PSC}$ during cold years in recent decades [WMO, 2011], with a new record occurring every few years, and it has been suggested that “the coldest Arctic winters are getting colder” [e.g., Rex et al., 2006], possibly as a consequence of climate change induced by increasing greenhouse gases.

More recent work, however, has questioned the statistical significance of the reported Arctic $V_{PSC}$ trends and offered new evidence that increases in well-mixed greenhouse gases (WMGHG) are unlikely to be the cause for the recently observed large Arctic stratospheric ozone losses [Rieder and Polvani, 2013, hereinafter RP13]. Furthermore, while it is well established that increases in WMGHG cool the upper stratosphere [Manabe and Wetherald, 1967; RP13; Sigmond et al., 2004], at the lower stratospheric levels relevant for ozone depletion, one finds little response to greenhouse gas forcing in chemistry-climate models.
Thus, whether the observed Arctic stratospheric cooling trends in cold years are caused by anthropogenic forcings of the climate system remains unclear, if the changes are indeed anthropogenic, then the key question is which forcing is the cause. For the polar stratosphere in the second half of the twentieth century, only two anthropogenic forcings are likely candidates: increasing concentrations of WMGHG [Trenberth et al., 2007] or increasing atmospheric burden of ODS [WMO, 2011]. The goal of this work, therefore, is to examine if and how these forcings have been affecting the Arctic wintertime stratosphere in recent decades.

Using observational and modeling data, we show that ODS are the dominant forcing of temperatures for the lower stratosphere (via decreases in the ozone concentrations), while WMGHG are the dominant forcing of temperatures in the upper stratosphere (via direct radiative cooling). The impact of ODS on temperature trends in the lower stratosphere is evident in the observations: in particular, we show that such trends have a clear seasonal cycle that directly connects them to ozone depletion and ultimately to changes in ODS. The signature of ODS is corroborated by the fact that these temperature trends have considerably decreased in the last decade, as ODS concentrations have leveled off as a consequence of the Montreal Protocol. This observational evidence is confirmed by the modeling evidence: when ODS and WMGHG are independently specified in a climate model with interactive stratospheric chemistry, their respective roles in controlling lower and upper stratospheric temperatures are immediately and unambiguously apparent, as demonstrated below.

2. Data and Methods

We analyze two widely used reanalysis data sets: the NASA-Modern Era Retrospective Analysis for Research and Applications (NASA-MERRA) [e.g., Rienecker et al., 2011] and the European Center for Medium-Range Weather Forecast (ECMWF) ERA-INTERIM reanalysis [Dee et al., 2011]. Since results for the two reanalysis are very similar, we show only NASA-MERRA in the body of the paper; results for ERA-INTERIM may be consulted for confirmation in the supporting information.

We focus on five quantities. The first two are temperature (\(T\)) and ozone mixing ratio (O3), which we consider at three different levels: 50 hPa, 30 hPa, and 10 hPa, chosen to roughly capture the lower, middle, and upper stratosphere, respectively (hereinafter referred to by the acronyms LS, MS, and US). We also consider the total ozone column (TOC), which is given in Dobson units. Finally, to relate our work with much literature that has been focused on PSCs, we also analyze the temperature extremes, by computing the area (\(A_{\text{PSC}}\)) and volume of air (\(V_{\text{PSC}}\)) cold enough for formation of nitric acid trihydrate clouds, i.e., with \(T < T_{\text{NAT}}\). Following Hanson and Mauersberger [1988] we take \(T_{\text{NAT}} = 195.59\) K at 50 hPa and \(T_{\text{NAT}} = 193.61\) K at 30 hPa. We compute \(V_{\text{PSC}}\) following the empirical formula

\[
V_{\text{PSC}} = 0.8 \times A_{\text{PSC}}(50 \text{ hPa}) \text{ km}^2 + 0.2 \times A_{\text{PSC}}(30 \text{ hPa}) \text{ km}^2 \times 5.06 \text{ km}
\]

of Rex et al. [2004].

Unless otherwise stated, by indicating a specific month, all quantities are means over the extended Arctic winter (i.e., January to April) and are averaged over the Arctic polar cap (i.e., 60°–90°N).

For the modeling component of this study, we analyze \(T\) and TOC from model integrations performed with the Canadian Middle-Atmosphere Model (CMAM) as part of the World Climate Research Program core project Stratospheric Processes and their Role in Climate (SPARC) Chemistry-Climate Model Validation Activity 2 (CCMVal-2). We analyze three sets of CMAM simulations for the period 1960–2099: (1) the SCN-B2c runs, with only ODS varying as per the WMO [2007] A1 scenario; (2) the SCN-B2b runs, with only WMGHG varying as per the SRES A1B scenario; and (3) the REF-B2 runs, a combination of (1) and (2); see the SPARC Report [SPARC-CCMVal, 2010] for further details on the scenarios and McLandress et al. [2010; 2011] for further details on the CMAM model.

3. Evidence From Reanalyses

We start by examining the notion that \(V_{\text{PSC}}\) (or \(A_{\text{PSC}}\)) is an exceptionally good proxy for ozone changes in the Arctic. In Figure 1 we compare scatterplots of ozone and \(V_{\text{PSC}}\) (or \(A_{\text{PSC}}\)) with scatterplots of ozone and polar cap \(T\) at 50 hPa: the results suggest that the latter quantity is a much better proxy for ozone. For the winter mean, this is clearly seen in Figures 1a–1d: Figures 1a and 1b show very high correlations—in excess
of 0.75—between T at 50 hPa and either O3 at that level (Figure 1a) or TOC (Figure 1b). When only the extreme temperatures (i.e., APSC and VPSC) are used (Figures 1c and 1d), the correlation becomes smaller (below 0.75).

Note that, in addition to being easier to calculate, T is more robust: APSC and VPSC are highly sensitive to the choice of a denitrification threshold temperature (TNAT), the horizontal resolution of the data (needed to compute an area), and the well-known biases present in individual reanalyses [e.g., RP13; Manney et al., 2003].

More important yet, the simple polar cap T appears to be a superior metric because its correlation with ozone does not collapse in March, the month when recent observed T trends are the largest. This is illustrated in Figures 1e–1h: note how the visual correlations disappear in the APSC and VPSC plots (Figures 1g and 1h), whereas they remain quite high for the T plots (Figures 1e and 1f). In Figures 1a–1h the colored dots indicate the few extremely cold winters that have occurred during the observational period (see the caption for details). If those extreme winters are removed from the analysis, the correlation in each panel is reduced (contrast correlation in red and black at the bottom of each panel). However, the reduction is much stronger for the panels using APSC or VPSC than for those using T. For completeness we show the results for the 30 hPa level in the supporting information (see Figure S1). Note that the connections are generally weaker at this level, not surprisingly as the bulk of ozone is found in the LS. Overall, the message from Figure 1 is clear: polar cap T is a better indicator of Arctic stratospheric conditions relevant for ozone changes than VPSC. We therefore adopt it for the remainder of this study.

Armed with this information, we next analyze recent Arctic trends, starting with ozone. In Figure 2 we plot time series of wintertime polar cap O3 at three different levels, together with the TOC time series at the bottom. It is noteworthy that in the LS and MS (Figures 2a and 2c), a strong and statistically significant negative trend is observed from 1980 to 2000. The fact that the ozone stops decreasing beyond the year 2000 strongly supports the view that the ozone trends represent the well-known signal of Arctic ozone depletion, caused by increasing ODS [WMO, 2011], after 2000 the Montreal Protocol began to reduce the atmospheric burden of ODS. For convenience, the time periods used and the computed trends (with significance levels) are shown at the bottom of each panel of Figure 2; they are visually indicated with black lines. Note that in the TOC curves particularly cold winters (such as 1996/1997 or 2010/2011) and the volcanic eruption of Mount Pinatubo in 1991 can be clearly seen.

Figure 1. Scatterplots of mean wintertime (January–April) polar cap (60°–90°N) (a) ozone mixing ratio (O3) and temperature (T), (b) total column ozone (TOC) and T, (c) O3 and area of polar stratospheric clouds (APSC), and (d) TOC and volume of polar stratospheric clouds (VPSC). (e–h) Same as Figures 1a–1d but for March only. All panels for NASA-MERRA in 1980–2013. All quantities at 50 hPa except TOC and VPSC. Colored dots mark particularly cold winters during the observational period: 1995/1996 (magenta), 1996/1997 (purple), 1999/2000 (green), 2004/2005 (orange), and 2010/2011 (red). Solid black lines mark the linear fits between the scatterplot quantities. The corresponding correlation coefficients for all years (black) and for data without the five particular cold winters (red) are reported in each panel.
In spite of the large interannual variability, the connection between ODS and LS/MS ozone is also clear from the fact that the TOC time series closely mirrors the time series of equivalent effective stratospheric chlorine (EESC), which we show (inverted) with the dashed blue curve in Figure 2g together with the TOC time series. We also note that the ODS/ozone connection is not obvious in the US (Figure 2e), for reasons that will become apparent below. Recall, however, that the bulk of the ozone layer is located around 50 hPa, explaining why the ODS/TOC connection remains clear. Next we examine whether these observed ozone trends are a constant feature throughout all winter months or if individual months are dominating the signal. Perhaps not surprisingly, we find that the observed trends are largely a springtime phenomenon, as one can see from the March time series shown in Figures 2b, 2d, 2f, and 2h.

Figure 2. Time series of mean polar cap (60°–90°N) ozone mixing ratios (O_3) in 1980–2013 from NASA-MERRA in (a) winter (January–April) at the 50 hPa level. (b) Same as Figure 2a but for March only. (c and d) Same as Figures 2a and 2b but for the 30 hPa level. (e and f) Same as Figures 2a and 2b but for the 10 hPa level. (g and h) Same as Figures 2a and 2b but for total column ozone (TOC). Solid lines represent statistically significant trends (significance level 95% or higher). Evaluation periods for trends are 1980–2000 in Figures 2a–2d, 2g, and 2h. These periods have been chosen to highlight the importance of ODS for Arctic ozone losses. For convenience the trends in kg kg\(^{-1}\) yr\(^{-1}\) in Figures 2a–2d and Dobson units (DU) yr\(^{-1}\) in Figures 2g and 2h and corresponding significance levels (in percent) marked with ‘at’ symbol are given in each panel. Blue hashed line in Figures 2g and 2h gives inverted EESC from the WMO [2007] A1 scenario for 1980–2013.

In spite of the large interannual variability, the connection between ODS and LS/MS ozone is also clear from the fact that the TOC time series closely mirrors the time series of equivalent effective stratospheric chlorine (EESC), which we show (inverted) with the dashed blue curve in Figure 2g together with the TOC time series. We also note that the ODS/ozone connection is not obvious in the US (Figure 2e), for reasons that will become apparent below. Recall, however, that the bulk of the ozone layer is located around 50 hPa, explaining why the ODS/TOC connection remains clear. Next we examine whether these observed ozone trends are a constant feature throughout all winter months or if individual months are dominating the signal. Perhaps not surprisingly, we find that the observed trends are largely a springtime phenomenon, as one can see from the March time series shown in Figures 2b, 2d, 2f, and 2h. Indeed, in that month, the ODS/ozone connection is even clearer than for the mean winter case (except for Figure 2f, which is discussed below); the other months, which show very little trends, are plotted in Figure S2. The fact that the ozone
trends occur largely in the spring should come as no surprise: it is well known that ozone depletion is only effective in the presence of sunlight, and the Arctic is dark until March. We note, in passing, that nearly identical results are obtained if ERA-INTERIM ozone is used (see Figures S3 and S4); hence, these results are not dependent on a specific analysis.

Given the statistically significant trends linking ODS increases to stratospheric ozone trends, it is now natural to ask: is it possible that these ozone trends could be the drivers of Arctic stratospheric temperature trends? To answer this, we plot 7 trends from the NASA-MERRA reanalysis in Figure 3; winter means in Figures 3a, 3c, and 3e; and March only in Figures 3b, 3d, and 3f, at the same three levels as Figures 2a–2f (again, we simply note that ERA-INTERIM yields very similar plots, given in Figure S5, for completeness).

Figure 3 shows, first, that in the LS and MS (Figures 3a–3d), the temperature time series map well onto the corresponding ozone time series in Figure 2. Second, contrasting Figures 3a, 3c, and, 3e and Figures 3b, 3d, and 3f, one can see that the observed Arctic temperature trends are again a spring time phenomenon; the other months (shown in Figures S6 and S7) show little or no trends. Third, focusing on the period with increasing ODS (1980–2000), we find cooling tendencies (i.e., significance level between 90% and 95%) for the winter mean in both the LS and MS (the dotted black lines in Figures 3a and 3c), and an even stronger signal in March, where statistically significant trends (i.e., significance level 95% or higher) can be seen (the
solid black lines in Figures 3b and 3d). Therefore, in spite of the large interannual variability that has been the focus of much recent literature, a clear cooling signal emerges in the lower and middle stratosphere when \( T \) as opposed to \( V_{PSC} \) is used; moreover, note that this cooling appears to vanish after the year 2000, in tandem with the ozone trends of Figure 2, which one would expect if ODS, via ozone depletion, are the main drivers of that cooling, as we suggest.

At this point we draw the reader’s attention to Figures 3e and 3f: in the upper stratosphere, there appears to be no slowing down of the cooling trend after the year 2000, in contrast to lower altitudes. In fact, there is a statistically significant trend all the way to 2013, with no hint of a kink around the year 2000, as in the ODS and \( O_3 \) curves. This is a clear indication that a different physical process is at play in the US: at those levels WMGHG are controlling the temperatures [e.g., Shine et al., 2003] and since the WMGHG increase is monotonic from 1980 to 2013, the temperature trends in the US are monotonic too. In fact, we suggest the causality link at those levels is opposite to the one in the LS and MS: the WMGHG induced cooling causes ozone to rise in the US. However, prior to 2000, the WMGHG signal is overwhelmed by the ODS forcing, resulting in ozone loss even at those levels; beyond 2000, however, the WMGHG forcing becomes dominant, and as a consequence, ozone starts to increase in the US. A hint of this can be seen in Figure 2e (and, less clearly, in Figure 2f). Of course, this is only a suggestion at this stage, since the two forcings cannot be separated in the observational data. To corroborate this suggestion, we now turn to modeling, where one can independently examine the effect of each forcing.

### 4. Evidence From Chemistry-Climate Model Integrations

We next examine three sets of integrations performed with the CMAM model. It is important to emphasize that in CMAM, the ozone concentrations are computed interactively and are therefore directly affected by the temperatures, in addition to affecting the temperatures themselves. In the three sets, each of which comprises three model runs and which cover the period 1960 to 2099, the forcings are as follows: in the first set only ODS vary in time, in the second only WMGHG vary, and in the third both vary. We note that no other forcing (e.g., volcanic aerosols, and the solar constant) is included in these runs. Also, these CMAM runs are performed with fully coupled ocean and sea ice components, so that no other external forcing beyond ODS and WMGHG is needed: this makes the interpretation of the computed ozone and temperature trends totally unambiguous. For further details, the reader is referred to McLandress et al. [2010; 2011].

The results for all three sets of CMAM integrations are summarized in Figure 4. Figures 4a–4c show the forcings: only ODS varying in Figure 4a; only WMGHG in Figure 4b; and both in Figure 4c. Note that EESC trends (blue lines) have opposite signs before and after the year 2000 (because of the Montreal Protocol), whereas the \( CO_2 \) concentrations (red lines) increase monotonically (following the IPCC SRES A1B scenario here, as prescribed by the CCMVal-2 project). The time series of modeled LS, MS, and US temperatures (Figures 4d–4m) and TOC (Figures 4n–4p) for the three different forcings are shown in the corresponding columns. In each panel the grey lines give the ensemble mean of a set of three runs, and the red lines show the corresponding 5 year running mean.

The left column in Figure 4 (panels 4a, 4d, 4g, 4k, and 4n) shows the runs with only time-varying ODS and WMGHG fixed at 1960 levels. As seen in Figures 4d and 4g, the model shows significant cooling trends in the LS and MS, of about \(-0.3\) to \(-0.35\) K/decade (respectively), over the period 1960–2000: these temperature trends must be caused by increasing ODS, since no other forcing is time varying in the model. What happens is this: the ODS cause ozone loss over that period (Figure 4n), which results in reduced UV absorption at those levels and therefore causes cooling trends at the height where most ozone loss is found (i.e., in the LS/MS). Interestingly enough, all of these trends reverse after the ODS turnaround, and from 2000 to 2099 we find a significant warming trend of about \(+0.11\) to \(+0.12\) K/decade at these levels as ODS concentrations slowly but steadily decline. In contrast, we find no significant trends or tendencies in the US (Figure 4k), as one would expect from the fact that WMGHG are fixed at 1960 levels in these runs and that both ODS and ozone losses are relatively small at 10 hPa.

Contrast these results with those in in the middle column of Figure 4 (panels 4b, 4e, 4h, 4l, and 4o) which show the runs with ODS fixed at 1960 levels but with monotonically increasing WMGHG (note that \( CO_2 \) concentrations more than double over the length of these runs). As documented previously in RP13, the LS
Figure 4. Temporal evolution of CO₂ (red) and equivalent effective stratospheric chlorine (EESC, blue) in (a) the SCN-B2c scenario (only ODS vary), (b) the SCN-B2b scenario (only WMGHG vary), and (c) the REF-B2 scenario (both ODS and WMGHG vary). (d) Mean wintertime (January–April) polar cap (60°–90°N) temperature (T) in 1960–2099 from the Canadian Middle- Atmosphere Model (CMAM) with only ODS varying. (e) Same as Figure 4d but with only WMGHG varying. (f) Same as Figure 4d but with both forcings varying. (g–i) Same as Figures 4d–4f but at the 30 hPa level. (k–m) Same as Figures 4d–4f but at the 10 hPa level. (n) Mean wintertime total column ozone (TOC) in 1960–2099 with only ODS varying. (o) Same as Figure 4n but with only WMGHG varying. (p) Same as Figure 4n with both forcings varying. Dotted lines in Figures 4a–4c mark the overlap with the observational period of the reanalyses data sets (1980–2013). Blue hashed boxes in Figures 4a and 4c mark the historical period (1960–2000) with an almost linear increase in EESC. Grey curves in Figures 4d–4p give the three-ensemble member mean while red curves give the corresponding 5 year running mean. Black solid lines in Figures 4d–4p give linear trend estimates (significance level 95% or higher). For convenience trends in K/decade in Figures 4d–4m and DU/decade in Figures 4n–4p and corresponding significance levels (in percent) marked with ‘at’ symbol are reported in each panel.
is largely unaffected by the WMGHG trends (Figure 4e), whereas the MS and US exhibit very clear and statistically significant cooling trends (Figures 4h and 4l): these temperature trends cause large ozone increases at these altitudes (see the TOC in Figure 4o), as a consequence of the temperature dependence of the gas phase chemistry of stratospheric ozone [e.g., Haigh and Pyle, 1982; Rosenfield and Douglass, 1998].

Third, consider the right column of Figure 4 (panels 4c, 4f, 4i, 4m, and 4p), where both ODS and WMGHG vary in time. The results for the combined ODS and WMGHG forcing runs are, largely, a superposition of the left (panels 4a, 4d, 4g, 4k, and 4n) and middle column (panels 4b, 4e, 4h, 4l, and 4o) of Figure 4: however, we can now better understand how the two anthropogenic forcings are able to affect the temperatures at different levels. In the LS (Figure 4f), temperatures are largely controlled by ODS (via ozone depletion) and clearly resemble those in Figure 4d. Conversely, in the US (Figure 4m), temperatures are largely controlled by WMGHG and clearly resemble those in Figure 4l. In the MS (Figure 4i), both forcings are able to affect the temperature; before 2000 their effects are additive, resulting in a statistically significant cooling trend, whereas in the future the two forcings largely cancel, resulting in statistically insignificant temperature trends. The temporal evolution of TOC in these CMAM integrations is therefore clear: as seen in Figure 4p, TOC declines before 2000 due to increasing ODS and thereafter rises mostly as a consequence of increasing WMGHG, whose impact is much larger than the impact of declining ODS (contrast Figures 4n and 4o from 2000 to 2099).

Finally, we ask whether these trends in CMAM are constant throughout the winter or whether any month dominates the trends in temperatures and TOC. As for the reanalysis data discussed in the previous section, we find that spring, particularly March (and to some extent April), dominates the modeled changes in both column ozone and LS/MS temperatures (see Figure S8). Hence, the CMAM model is able to capture the seasonality of the observed trends there, in both the only-ODS and all forcings cases. This adds confidence to our interpretation of the roles of the different anthropogenic forcings on the observed trends.

Before concluding, one small caveat should be noted. ODS are also greenhouse gases [e.g., Trenberth et al., 2007] and hence have a direct radiative effect on temperature. Given the model output available to us, we cannot isolate this direct effect from the indirect one (via ozone depletion). Nevertheless, the direct effect is likely to be small in the lower stratosphere, as can be seen by contrasting the results from the only ODS and both runs.

5. Conclusions

The first conclusion of our study is that temperature extremes, commonly used to evaluate the area or volume of PSCs, are not optimal metrics for analyzing past and future ozone and temperature trends in the Arctic. Instead of focusing on $A_{PSC}$ or $V_{PSC}$, we have shown that a simpler and more robust quantity—the polar cap mean temperature—is useful for understanding recent Arctic stratospheric trends in ozone, not only averaged over the winter but also in the key month of March.

This understanding yields the second contribution of our study. We have shown that both ODS and WMGHG play key roles in the Arctic stratosphere, but they affect different levels and in different ways. ODS control the lower stratosphere (via ozone depletion), whereas WMGHG control the upper stratosphere; the middle stratosphere is affected by both. Furthermore, ODS affect temperatures by first changing the ozone concentration, whereas WMGHG affect the temperature first which then affects the ozone concentration: hence, the ozone/temperature causality link is opposite for these two anthropogenic forcings. As a consequence of this relationship, it seems likely that the recently observed Arctic temperature trends have been caused by ODS (via ozone depletion), whereas WMGHG will play a bigger role in coming years as the stratosphere cools due to their projected increase. Nonetheless, as ODS concentrations are projected to decrease rather slowly in coming decades and as dynamical processes are known to also influence the polar stratosphere [e.g., Braesicke and Pyle, 2003; Pyle et al., 2005; Shepherd, 2008], it is entirely possible that individual cold years and large ozone losses might still occur.

References


