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NEW DIRECTIONS

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New Directions: Stratospheric ozone recovery in a changing atmosphere ☆

About 90% of the ozone in the Earth's atmosphere resides in the stratosphere, forming the "ozone layer" which shields life on Earth from harmful ultraviolet radiation. In the mid-1970s, it was recognized that anthropogenic chlorofluorocarbons (CFCs) could deplete the ozone layer. Observations of the ozone layer indicated that depletion was occurring due to the buildup of CFCs and Halons in the stratosphere. The most severe depletion was discovered over the Antarctic in the mid-1980s, which is commonly referred to as the "ozone hole". To protect the ozone layer, the Montreal Protocol was crafted in 1987 by the United Nations to address this global problem. Fig. 1 shows the predicted trends in the stratospheric chlorine concentration as specified by the Montreal Protocol and its subsequent amendments.

Many observations in the stratosphere indicate that the total chlorine abundance is at or near a peak, in agreement with the predictions shown in Fig. 1. Future ozone recovery is primarily linked to halogen loading in the stratosphere. Assuming global compliance with the Montreal Protocol, the ozone layer is expected to recover by the middle of the 21st century (Fig. 2). Time-dependent data and model simulations of ozone are shown in Fig. 2. The range of model predictions come from various models that utilize different assumptions about the future climate and composition of the atmosphere. These assumptions account for estimated differences in atmospheric composition between 1980, before the ozone hole began, and 2050. The 1992 Pinatubo volcanic aerosols caused the drop in the measured and modeled ozone concentration seen in Fig. 2. Cloud and aerosol surfaces increase the ozone depletion potential of reactive halogen and nitrogen gases through heterogeneous chemistry. An outcome not included in models is the occurrence of one or more large eruptions in the coming decades. Such events can delay ozone recovery for several years following each eruption.

In addition to chemical changes, observational data indicate that the global climate in the stratosphere has cooled over the last two decades. The magnitude of cooling is approximately 1 and 2 K in the lower and upper stratosphere, respectively. This cooling is attributed to ozone depletion in the stratosphere as well as increases in well-mixed greenhouse gases (GHGs) including water vapor. Climate change in the stratosphere will affect the rate of ozone recovery differently at various altitudes and latitudes. Stratospheric cooling is predicted to increase the abundance of ozone in the upper stratosphere because the rates of chemical reactions that deplete ozone decrease with decreasing temperatures. Thus, this effect in isolation can substantially hasten the predicted rate of ozone recovery. However, large uncertainties exist as to how climate change will affect the rate of ozone recovery in the lower stratosphere. These uncertainties reflect the range of model predictions shown in Fig. 2.

Unlike the upper stratosphere, sulfate aerosols are abundant in the lower stratosphere. Enhanced heterogeneous chemistry on sulfate particles in a future colder and more humid lower stratosphere can cause more global ozone depletion. Similarly, enhanced heterogeneous processing in the polar regions, leading to more ozone losses there, can decrease mid-latitude ozone levels through transport and mixing of air masses. Although chemical causes of ozone depletion are relatively well known, dynamical factors, which play an important role in controlling the ozone abundance in the lower stratosphere, are not well characterized in coupled chemistry-transport models. Some studies indicate that the observed decreases in northern hemisphere midlatitude ozone over the last two decades must have a strong dynamical component. Currently models, when compared against observations, underestimate the "mean age" of air in the stratosphere, indicating that transport timescales are not well simulated. Ongoing and future work to resolve such discrepancies can help to better quantify how dynamical changes will affect the rate of ozone recovery. If the past dynamical changes are due to natural variability, then ozone recovery could potentially be hastened or delayed due to natural changes in the dynamics of the stratosphere. On the

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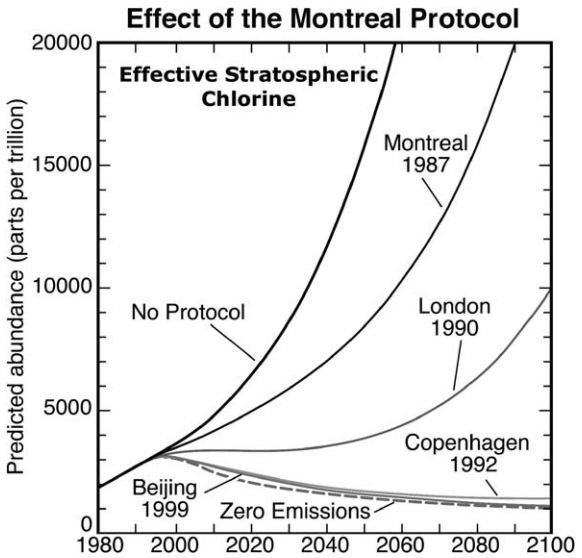

Fig. 1. Predictions for the future abundance of effective stratospheric chlorine assuming (1) no Protocol regulations, (2) only the regulations in the original 1987 Montreal Protocol and (3) additional regulations from subsequent amendments. The names of the city and year refer to the location where the changes to the original Protocol were agreed upon. The zero emission line indicates the effective stratospheric chlorine if all emissions were terminated in 2003.

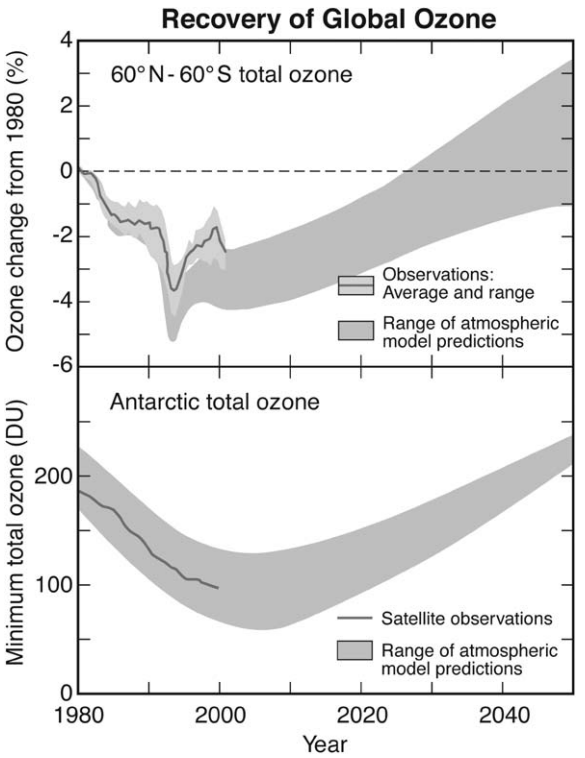

Fig. 2. Observed and predicted global total ozone (top panel) and minimum total ozone over Antarctica (bottom panel). In the top panel, the mean observed global ozone is shown in the solid line, while the range of observations is shown in the light shading between 1980 and 2002. The range of predictions from atmospheric models assuming different scenarios about the future climate and atmospheric composition is shown in the darker shading between 1980 and 2050. In the lower panel, satellite observations of the minimum total ozone over Antarctica is shown in the solid line, while the range of model predictions is shown with the shading. The column ozone concentration in the lower panel is given in Dobson Units (DU). One DU equals 2.7×10^{16} ozone molecules cm^{-2}.

other hand, if the past dynamical changes are mainly driven by GHG forcing, then it is likely for such changes to increase in magnitude in future decades, thus delaying the rate of ozone recovery.

The climate and chemistry in the stratosphere are influenced in part by the water vapor concentration. The water vapor mixing ratio in the lower stratosphere is currently increasing at a rate of ~ 0.5 ppm/decade. This rapid increase in lower stratospheric water vapor cannot be simply explained by methane oxidation. In addition to being a greenhouse gas, water vapor increases can elevate HO_x levels, affecting the NO_x and ClO_x cycles of ozone depletion in the stratosphere. Thus, to fully assess the rate of ozone recovery requires knowledge of stratospheric water vapor changes in future decades.

Originally, it was suggested that more water vapor is being transported into the stratosphere as increasing GHGs further warm the tropical tropopause, where the transport process occurs. During transport, a warmer tropopause can hold more water vapor before condensation occurs than a colder tropopause. Recent observations, however, indicate that the tropical tropopause is cooling, which implies less water vapor transport is occurring across this boundary. Another hypothesis states that strong tropical convection may be overshooting more ice particles directly into the stratosphere in recent decades. Water isotope data collected last year during the CRYSTAL-FACE field program in Florida may shed further light on this idea. Yet another theory suggests that the area of transport in the tropics may have expanded in recent decades, causing transport of more water vapor into the stratosphere, although currently, there is no observational data to refute or provide support for this hypothesis. Suffice is to say that the cause of this rapid increase is yet unknown and more research and field work is needed in the areas indicated above to resolve these issues. Until the causes of water vapor changes in the stratosphere are better understood, it will be difficult to assess their future impact on ozone recovery.

Another interesting aspect of ozone recovery that has been discussed in the literature over the last year is the effect of anthropogenic gases, other than CFCs and

Halons, on stratospheric chemistry. As halogen concentrations decline in the stratosphere (Fig. 1), the chemical cycles of ozone depletion will be influenced more by HO_x and NO_x cycles. Therefore, species that can elevate HO_x and NO_x levels in the stratosphere could, in principle, delay the recovery of stratospheric ozone. In a recent study, the predicted increase in stratospheric NO_x, mainly due to increased N_2O emissions, is shown to delay the ozone recovery in the next 50 years (Randeniya et al., 2002, Journal of Geophysical Research 29, doi:10.1029/2001GL014295). Another study raises concern about stratospheric ozone depletion in a future hydrogen fuel-cell economy (Tromp et al., 2003, Science 300, 1740). Hydrogen fuel cells are known to leak hydrogen to the atmosphere, potentially causing an increase in the abundance of hydrogen gas and subsequently stratospheric water vapor in future decades. As indicated above, increases in stratosphere water vapor can impact both the climate and chemistry of the stratosphere. The above studies highlight new research areas that will require further examination.

This discussion is incomplete without addressing a few points regarding ozone recovery in the polar regions. The springtime Antarctic "ozone hole" is expected to recover according to the timeline shown in Fig. 2. However, the recovery of Arctic ozone, particularly over the next couple of decades, while anthropogenic halogen levels are still sufficiently high (Fig. 1), is less predictable. Some studies show that the cooling climate in the stratosphere can increase Arctic cloudiness, resulting in denitrification. A denitrified Arctic stratosphere in early spring is primed for ozone destruction because reactive nitrogen that can mediate ozone loss, by sequestering active chlorine, has been removed from the lower stratosphere. A number of model simulations show that severe denitrification can increase springtime Arctic ozone losses by about 30%, causing a delay in the recovery of Arctic ozone. However, severe denitrification over a broad altitude range in the Arctic cannot be simply caused by GHG cooling alone. A major contribution from currently not-well-understood dynamical factors are needed to enhance the direct cooling effect of GHGs. Therefore, whether severe denitrification, over a broad altitude range, will occur in the Arctic before the full ozone recovery occurs remains an open question. If severe and deep denitrification does occur in the Arctic, then springtime column ozone losses, exceeding those measured in the 1990s, are possible in the next few decades.

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