### **Stratospheric Composition**

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Quantifying the anticipated response of the stratospheric ozone layer to anthropogenic changes in composition in the 21<sup>st</sup> century constitutes a key challenge for Earth System Science.

Photochemical and radiative processes affecting the stratospheric ozone layer couple the troposphere and stratosphere. The ozone layer absorbs ultraviolet (UV) radiation, protecting life on Earth by controlling how much UV radiation reaches the surface. Stratospheric ozone is produced by photolysis of oxygen molecules and destroyed chemically by catalytic processes involving trace gases containing nitrogen, hydrogen, and halogen (chlorine and bromine) atoms. The stratospheric circulation controls many aspects of the total ozone distribution.

Dynamic and photochemical processes both influence and are influenced by ozone layer evolution. Ozone heating affects the stratospheric circulation and ozone itself is a greenhouse gas (GHG) in the lower stratosphere and upper troposphere. Ozone evolution in the next century depends on both decreases in ozone depleting substances (ODSs) and GHG increases that alter stratospheric temperature and circulation.

Halogen-containing compounds increased by more than a factor of four between 1965 and 2000, causing a decrease in global stratospheric ozone and severe ozone depletion at high latitudes (e.g., the Antarctic ozone hole (AOH)). The AOH leads to high springtime surface UV and also forced trends in southern hemisphere tropospheric climate. The 1987 Montreal Protocol (MP) and its amendments regulate production of ODSs (chlorofluorocarbons and halons). Although ODS levels have been declining since the 1990s due to the MP, their atmospheric lifetimes are long (decades to centuries), and the current rate of ODS decrease is much smaller than its prior rate of increase.

The main GHGs (carbon dioxide ( $CO_2$ ), methane ( $CH_4$ ), and nitrous oxide ( $N_2O$ )) continue to increase. ODSs and their replacement compounds are also powerful GHGs; changes in GHGs alter stratospheric circulation and climate. Simulations show increased lifting in the tropical lower stratosphere that decreases the tropical ozone layer and alters distributions of ODSs and other GHGs.  $CO_2$  increases also cool the middle and upper stratosphere, slowing temperature dependent catalytic loss cycles and increasing upper stratospheric ozone.

#### This challenge is timely.

In models, the importance of ODS decrease relative to GHG increase will change in the 2020-2030 timeframe. Observations are necessary to expand our knowledge of the climate-stratospheric composition system and test the adequacy and reliability of models used to project future ozone levels.

The expected ozone increase due to ODS decrease is difficult to isolate in a backdrop of natural variability and GHG forced changes in circulation and climate. Although models agree that the mass entering the stratosphere will continue to increase, there is no consensus as to the vertical extent of the tropical circulation increase or the latitudinal distribution of increased downwelling. Consequently, future evolution of lower stratospheric midlatitude ozone is in question. Chemical and dynamical processes in the lower stratosphere will contribute to the change in total ozone column at midlatitudes, thus composition measurements are to quantify an ozone increase due to ODS decline apart from the influence of changes in circulation and climate. Such attribution is necessary to demonstrate the benefit of the MP.

The evolution of aerosols and stratospheric water vapor contribute to uncertainty in prediction of the stratospheric ozone and climate change. Stratospheric water vapor, a source of hydrogen radicals and an important GHG, increases as the tropical tropopause temperature increases (and vice versa). Increasing or decreasing water in the lower and lowermost stratosphere amplifies or opposes the impact of other GHGs increases through radiative processes. Stratospheric aerosols affect surface temperature, cirrus cloud formation in the upper troposphere/lower stratosphere, stratospheric dynamics and ozone loss processes (through heterogeneous chemical reactions on the particle surfaces).

## Global measurements are necessary.

Global measurements of ozone, temperature,  $H_2O$ , and related constituents are necessary to reduce uncertainty in projections of future ozone levels. The natural variability of the system must be quantified to identify signatures of anthropogenic change. Attribution of an ozone increase due to ODS decrease is necessary information for the parties to the MP. The midlatitude column ozone increase due to climate and circulation change, projected by the end of the century, is substantial and would decrease the amount of UV radiation reaching the surface compared with historical levels. Continuing observations are necessary to quantify the evolution of stratospheric aerosols as well as their impact on the ozone layer and the Earth radiation balance. Aerosol observations and analysis are at the foundation of proposed efforts to mitigate climate change through geo-engineering.

#### Existing and planned US and international programs

Previous and ongoing satellite missions have established sound observational techniques and have yielded important results:

- The Microwave Limb Sounder (MLS) and Ozone Monitoring Instrument, both on NASA's Aura satellite, provide global measurements of total column ozone and ozone profiles. MLS measurements of H<sub>2</sub>O, hydrogen chloride (HCl), chlorine monoxide (ClO), and N<sub>2</sub>O can be used to quantify dynamic and photochemical changes in ozone. Aura was launched in 2004 and is five years past design lifetime.
- The Atmospheric Chemistry Experiment (ACE) a Fourier transform infrared spectrometer on the Canadian SciSat (launched in 2003), provides profiles of many constituents through solar occultation.

- Global total ozone and ozone profiles will continue from the Ozone Mapping Profiler Suite (OMPS) on Suomi NPP and the Joint Polar Satellite System (JPSS-1) to be launched in 2017. Attribution of ozone change to chlorine change or quantifying the ozone change due to climate change is not possible without additional measurements. Stratospheric aerosols are obtained from Suomi NPP and will be obtained from JPSS-2 (expected launch 2022) but not JPSS-1.
- The Stratospheric Aerosol and Gas Experiment III (SAGE III) will measure profiles of aerosols, ozone, temperature, nitrogen dioxide ( $NO_2$ ) and  $H_2O$  from the upper troposphere through the stratosphere from the International Space Station (ISS), approximately 2016 2020.

There are no planned missions that would include measurements of trace gases other than ozone and water vapor during the time period when the balance of processes controlling ozone evolution is most uncertain (2020 – 2035). Existing technology, however, places the community in a good position for follow-on missions. An ACE-type instrument deployed on the ISS (proposed to the EV program by APEX) or timely deployment of a version of an Aura instrument (e.g., MLS or an infrared limb sounder) would provide the needed measurements. Details of suggested space-based, suborbital and ground-based measurements are given in Table 1.

# Linking space-based and ground based observations

Ground based observations play an important role. NOAA and AGAGE networks measurements of all of the major ODSs underlie predictions of emissions and the evolution of these source gases to the  $22^{nd}$  century. NDACC provides column measurements of key stratospheric gases. The multi-decadal NDACC timeseries complement existing datasets and continued measurements will ensure the success of future deployments of space-based instruments.

#### Scientific and societal benefits

Monitoring stratospheric ozone, and constituents such as HCl and  $N_2O$  necessary to quantify the ozone responses to ODS decline and to changes in circulation and climate due to GHG growth, is beneficial to society because stratospheric ozone controls the amount of UV light reaching the Earth's surface, thereby protecting life. Such observations are key to fulfilling responsibilities to the US Congress and to the Vienna Convention and the Montreal Protocol.

Table 1. Measurements to meet this Challenge										
Measurement:	Rationale	Sampling		Resolution		Precision				
Crucial ( <i>C</i> ), Important ( <i>I</i> ), Useful ( <i>U</i> )		Spatial	Temporal	(lat x lon)	Vertical					
Total Column Ozone - ( <i>C</i> )	Quantify column ozone response to ODS decrease and GHG increase	Global	Daily	1° x 2°	N/A	3 DU				
Ozone profiles – ( <i>C</i> )	a) Quantify stratospheric and tropospheric ozone trends b) Determine changes in tropical upwelling	Global	Daily	5° x 10°	2 km	0.1 ppmv				
Temperature Profiles – ( <i>C</i> )	a) Quantify ozone response to temperature change. b) Determine temperature trends	Global	Daily	5° x 10°	2 km	2K/ decade				
Solar flux (spectrally resolved) – (I)	Quantify ozone variability due to solar processes	Global input	Daily	N/A	N/A	10%/ decade/ 10 nm				
Sulfate aerosols – (I)	Quantify radiative and photochemical effects of the natural aerosol layer	Global	Daily	5° x 10°	2 km	10%/ decade				
H <sub>2</sub> O, N <sub>2</sub> O, CH <sub>4</sub> , HCl – ( <i>C</i> )	Attribute ozone change to ODS decrease	Global	Monthly	10° x 10°	4 km	20%/ decade				
NO <sub>2</sub> , ClONO <sub>2</sub> , ClO, BrO, OH, HO <sub>2</sub> - ( <i>U</i> ) [Aircraft, Balloon, & Ground-based]	Quantify relative balance of ozone loss processes as composition changes	Selected locations (campaigns)	>Annual		< 1 km, tropopause to 100 hPa	20%/ decade				

BrO (sub pptv), CH <sub>3</sub> Br, CHBr <sub>3</sub> , other VSLSs, Halons (aircraft) – ( <i>U</i> )	Determine processes controlling VSLS contribution to stratospheric bromine	Tropical locations (campaigns)	> Annual		< 1 km, 500 hPa to 100 hPa	
Surface measurements of CFCs and HCFs (C)	timeseries of ODSs and replacement compounds	Global	Monthly	Surface meas.		10%/ decade
H <sub>2</sub> O , HDO– ( <i>C</i> )	Quantify trends in stratospheric water and their controlling mechanisms	Global	Weekly	2° x 15°	2 km	10%/ decade
CH <sub>4</sub> , N <sub>2</sub> O, CO <sub>2</sub> , SF <sub>6</sub> , CFC- 11, CFC-12 – ( <i>C</i> )	Quantify variability and trends in stratospheric circulation	Global	Monthly	10° x 10°	2 km	10%/ decade
H <sub>2</sub> SO <sub>4</sub> aerosol (C), precursors SO <sub>2</sub> (C), OCS (I)	Determine stratospheric background aerosol source	Global (UT +Strat.)	Weekly to Monthly	2° x 5°	2 km	20%, 5-10 pptv, 20- 50 pptv