Title: Coupling Between Climate Forcing and Stratospheric Ozone Loss Over the US in Summer: UV Dosage Levels and Human Health

James G. Anderson and Steven C. Wofsy

I. ***Key Challenges for Earth System Science***

Summary of the Problem:

We now know, through the union of high altitude *in situ* aircraft, satellite and NEXRAD weather radar observations, that the stratosphere over the US in summer is vulnerable to ozone loss as a result of a series of factors unique, worldwide, to the central US in summer. The key elements that couple climate forcing to ozone loss in the lower stratosphere over the US in summer are summarized in Figure 1 and reviewed here:

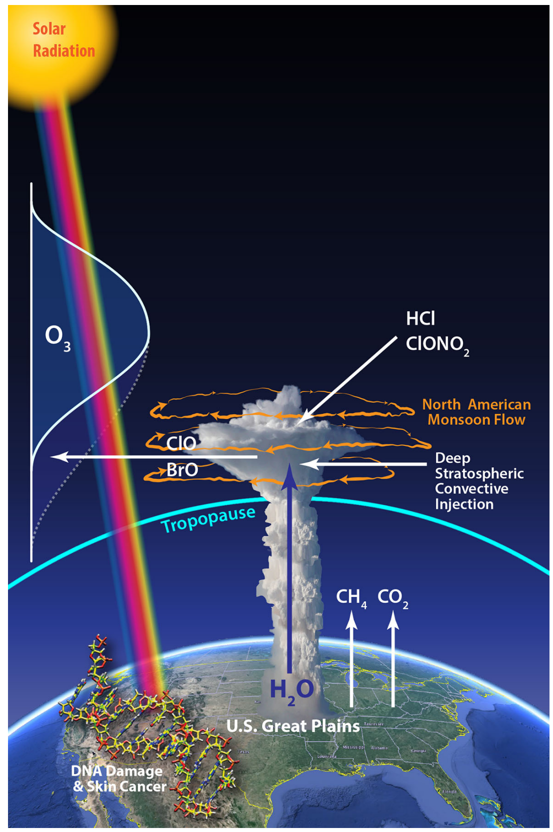

Figure 1. Graphical representation of the link between climate forcing by increased CO_2 and CH_4 release, convective injection of water deep into the summer stratosphere over the US, entrapment by the anticyclonic North American monsoon flow and the catalytic chemistry linking halogen radicals to ozone loss in the lower stratosphere.

1. Convection, as a result of severe storms over the Great Plains, delivers both markedly enhanced water vapor, as well as catalytic radical precursors from the lower troposphere, deep into the stratosphere over the US in summer.
2. The depth of convective injection is sufficient to reach altitudes of rapidly increasing available inorganic chlorine that is then catalytically converted on simple, ubiquitous, binary water-sulfate aerosols, to free radical form, ClO.
3. It is the chlorine radical, ClO, that couples to the available BrO radical to form a catalytic cycle rate limited by $ClO + BrO \rightarrow Cl + Br + O_2$ in combination with $ClO + ClO \rightarrow ClOOCl$ that constitutes the catalytic mechanisms capable of removing ozone; and it is the same mechanism that contributes to very large ozone loss over the polar regions in winter.
4. Anti-cyclonic flow in the lower stratosphere over the US in summer, as a result of the North American monsoon, contains that convective injection in a gyre that provides time (days to weeks), for photochemistry to catalytically remove ozone.
5. The remarkable sensitivity of increases in the skin cancer incidence in the US to fractional decreases in ozone column concentration places strict demands on the quantitative understanding required to accurately *forecast* ozone losses and the requisite increase in skin cancer incidence in the US in a changing climate.
6. The frequency and intensity of severe storms over the Great Plains of the US is increasingly tied to increased forcing of the climate by increasing levels of CO_2, CH_4 and N_2O in the Earth's atmosphere. As a result, climate forcing is mechanistically linked to forecasts of ozone reduction in the stratosphere over the US in summer.
7. These issues of the close coupling between heterogeneous and homogeneous catalytic ozone loss with the associated aerosols in the lower stratosphere must be linked to emerging strategic plans for climate engineering via solar radiation management (SRM) that proposes the addition of aerosols to the lower stratosphere to reflect solar radiation, thereby reducing shortwave forcing of the climate.

These developments engage the advancing union of observational techniques, the human health implications of ozone loss, the issue of proposed climate engineering via SRM, and key evidence of the climate structure from the paleorecord.

II. Key Unanswered Scientific Questions in the Coupling of Climate Forcing with Ozone Loss in the Stratosphere Over the US in Summer

We can summarize the factors that have emerged regarding (a) the dynamical structure of the stratosphere over the US in summer and (b) the heterogeneous and homogeneous catalytic chemistry of the lower stratosphere that, taken together, converge to imply that the column concentration of ozone in July and August is vulnerable to reductions involving the same reactions responsible for ozone loss over the Arctic in winter. The mechanisms that link climate change with ozone loss in the stratosphere are summarized in Figure 2.

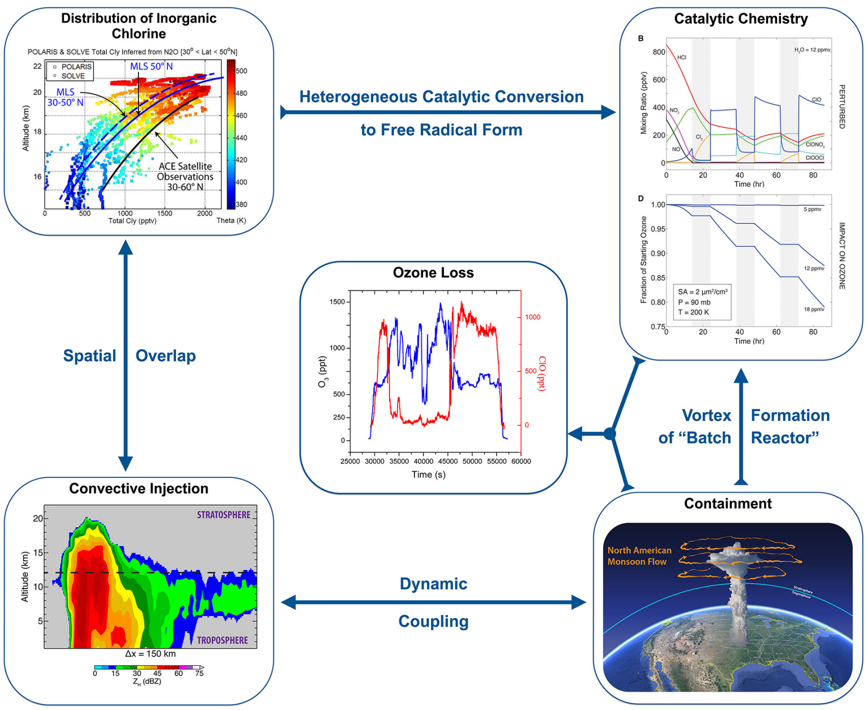

Figure 2. There are unique aspects of the stratosphere over the US in summer that conspire to produce the situation wherein the ozone column is vulnerable to reduction resulting from the combination of convective injection, containment by the anti-cyclonic flow in the lower stratosphere and catalytic photochemistry resulting from the conversion of inorganic chlorine to free radical form in simple binary sulfate-water aerosols.

The recognition of the unique attributes of the dynamics and catalytic chemistry of the lower stratosphere over the US in summer creates the imperative to answer the following questions.

1. What combination of physical mechanisms is responsible for the observed deep stratospheric convective injection of water vapor into the stratosphere over the US in summer?
2. How will the processes that compel that deep stratospheric injection respond to increased forcing of the climate by the addition of increasing amounts of CO_2, CH_4, N_2O, etc. to the atmosphere?
3. What is the structure of the 3D velocity fields within the storm structure in the stratosphere? What factors control the degree of irreversible injection of water and radical precursors into the stratosphere?
4. What is the structure of the wave breaking events associated with the convective intrusion? How do these wave breaking events affect the vertical kinetic energy of the deep convective events?

5. What are the concentrations of species co-injected with water by the convective event?
6. What are the concentrations of (a) the reactive chemical precursors that are normally removed by photolysis or oxidation in the troposphere and (b) the chemical tracers brought in by the convective storm or drawn in by the perturbation associated with the convection?
7. How does the structure of the convective injection alter the potential temperature surfaces in the vicinity of the injection?
8. What is the subsequent fate of the chemical cocktail convectively injected into the stratosphere in the hours and days following the convective injection?
9. Does the catalytic photochemistry linking sulfate aerosols and water vapor to the conversion of inorganic chlorine to free radical form mirror the same chemistry as a function of temperature and water vapor that was verified in the stratosphere over the Arctic?
10. Does the region of convective injection into the lower stratosphere decrease in temperature as a function of time due to enhanced radiative cooling in agreement with calculated cooling rates?
11. What is the observed covariance of the local ozone concentration with the rate limiting free radicals ClO and BrO as a function of time following convective injection?
12. What combination of observables are best suited for testing the response of stratospheric ozone to increasing convection into the stratosphere? What is required to develop a trusted forecast of ozone column concentrations over the US in summer in the face of increased climate forcing by CO_2 and CH_4.

III. Why is This Issue Timely:

- The ozone shield is arguably the most delicate aspect of habitability on the planet's surface. If the entire stratospheric ozone column were brought to standard temperature and pressure, that ozone column would be ~ 0.5 cm in depth. A 5% reduction in column ozone concentration over the US translates to a 15% increase in new skin cancer cases each year — i. e. slightly exceeding 500,000 additional skin cancer cases per annum in the US alone. This defines the accuracy to which the forecast of ozone column concentration changes must be quantitatively trusted in response to increased forcing of the climate by CO_2, CH_4, N_2O, etc.
- We have experience with, and have witnessed directly, the dramatic loss of ozone within the polar vortices in both hemispheres in late winter and early spring each year. The scientific community has established the detailed mechanistic link between the available inorganic chlorine in the stratosphere and the catalytic conversion of that inorganic chlorine to free radical form on simple, ubiquitous sulfate-water aerosols in the lower stratosphere. With the combination of *in situ* aircraft, satellite and NEXRAD observations, we now know that the conditions required for catalytic removal of ozone can be met over the US in summer as a result of deep stratospheric convective injection of water vapor and radical precursors in combination with extended containment of that injection by the anticyclonic flow in the lower stratosphere over the continental US resulting from the North American monsoon in July and August.
- The dependence of the frequency and intensity of severe storms over the US is increasingly tied in the scientific literature to the increased forcing of the climate by the release of greenhouse gases from fossil fuel combustion. Given the millennial scale lifetime of carbon dioxide in the atmosphere, the response of the climate system is irreversible on the decadal time scale of importance to society. It follows that so too are reductions in ozone column concentration and requisite increases in UV dosage levels irreversible with increased forcing of the climate.

IV. How are Space-Based Observations Linked to Suborbital and Ground-Based Observations

Figure 3 graphically displays the combination of space-based and suborbital observing systems that are required to address the problem of ozone loss over the US in summer. The observational strategy combines long-duration balloon capability, stratospheric solar propulsion for horizontal navigation in the stratosphere, and stratospheric reel-down capability for long-extension vertical soundings providing *in situ* detection of radicals, isotopes, ozone, reactive intermediates, long-lived tracers and condensed and vapor phase H_2O and HDO, (b) NEXRAD storm tracking with the capability to connect severe storm formation in the troposphere with convective penetration into the stratosphere, and (c) a new generation of small satellites that builds on the current NASA A-Train satellites. This coupled array of observations extends the capability of the approach from temporally sustained, high resolution (10 meter) *in situ* observations, to medium resolution (100 meter) linked *in situ* and remote observations, to regional (10 – 100 km) resolution coupled observations to finally global coverage. But with the observations linked to a common coordinate system by a combination of high accuracy tracers and simultaneous, spatially registered, data sets.

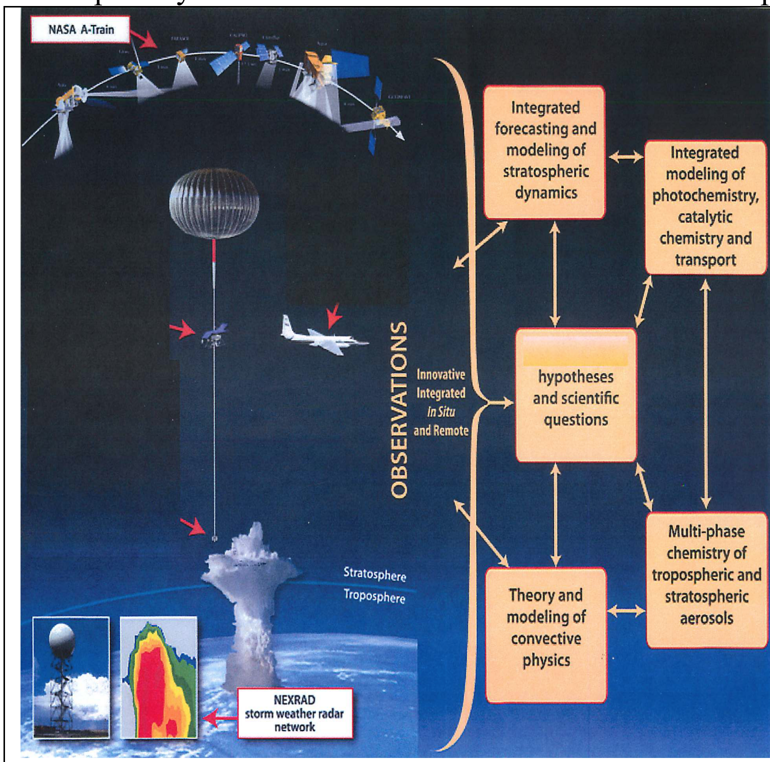

Figure 3. Ozone loss over the US in summer introduces a new observing system to the Earth sciences to quantitatively address critical issues linking increasing climate forcing with climate response involving severe storms, ozone loss and climate feedbacks. Observations that provide a quantitative coupling between repeated high resolution, *in situ*, observations employing a long duration StratoCruiser system, the NEXRAD network of radars and the NASA A-Train satellites.