

Analysis of Convectively Injected Water Vapor and Potential Impact on Chlorine and Ozone Over the US in Summer

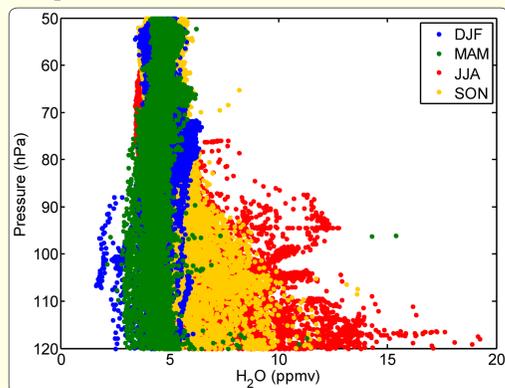
David Wilmouth, Jessica Smith, John Dykema, and James Anderson

Department of Chemistry and Chemical Biology & School of Engineering and Applied Sciences, Harvard University, Cambridge, MA

Introduction

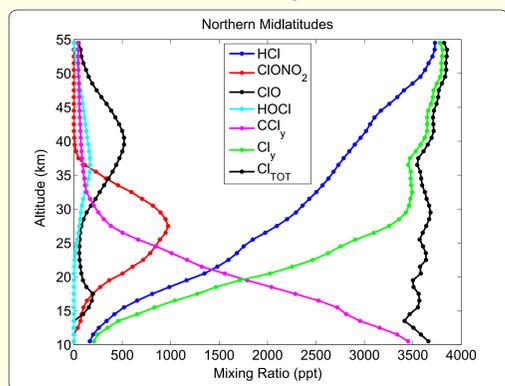
Elevated water vapor concentrations from convective injection have been observed repeatedly in the lower stratosphere over the United States during the summertime. Here we examine the in situ aircraft and satellite measurement data record of high water and analyze in depth a case study for August 26-31, 2013 relevant to the SEAC⁴RS mission. We further explore the potential for elevated stratospheric water vapor to impact inorganic chlorine and ozone as a function of altitude and highlight the most significant factors in the model calculation. Particular focus is given to a scenario with elevated aerosol surface area.

In Situ H₂O Observations over the US in Summer



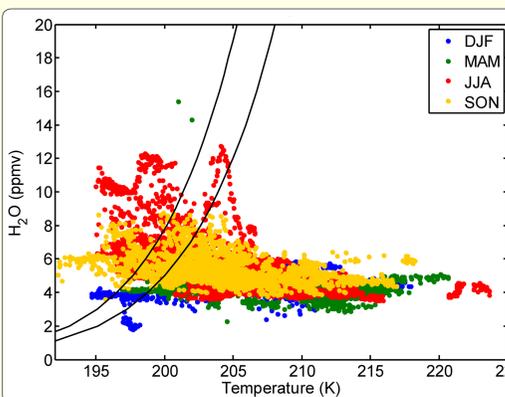
In situ aircraft measurements of water vapor in the summertime over the United States show numerous occurrences of elevated concentrations reaching pressure altitudes deep into the stratosphere.

Vertical Profiles of Chlorine Species



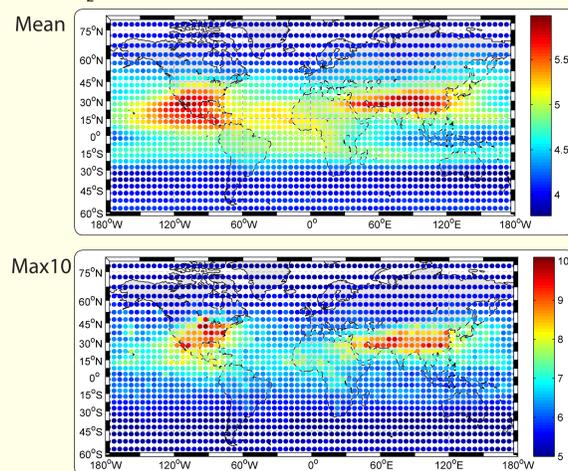
Average stratospheric chlorine for northern midlatitudes (30–60°) from 2004 ACE-FTS satellite measurements (Nassar et al., JGR 2006). Heterogeneous reactions, which are highly dependent on temperature and water vapor, can repartition inorganic chlorine from HCl and ClONO₂ into more photolabile forms and sequester nitrogen oxides.

Water and Temperature Observations at 90 ± 10 mb



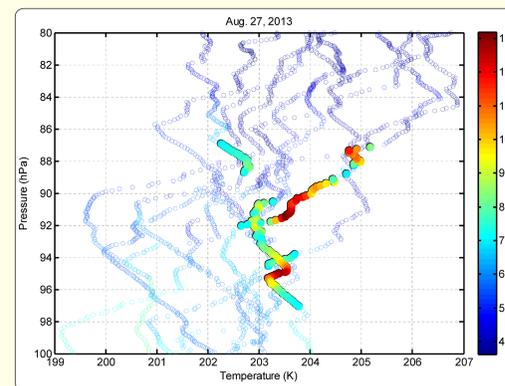
In situ observations of water vapor mixing ratios and temperatures at 90 ± 10 mb, color-coded by months, superimposed on a plot of the threshold conditions for chlorine activation at aerosol surface areas of 2 and 10 μm²/cm³.

MLS H₂O Observations in Summer at 100 mb



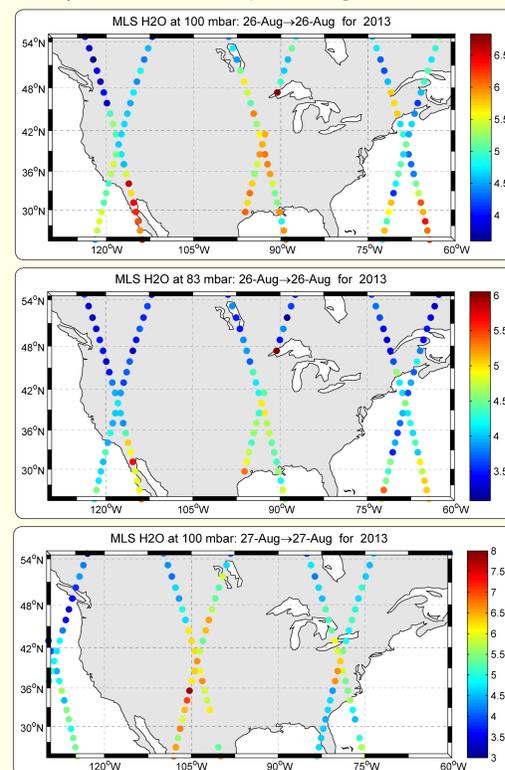
Aura MLS data July-August 2005-2013. The highest water vapor at the highest latitudes anywhere on the globe is over the continental US in summer.

Case Study: in Situ Water Vapor - Aug 27, 2013



Water vapor in excess of 13 ppm at ~410 K was observed in situ on-board the ER-2 during the SEAC⁴RS flight of Aug 27, 2013.

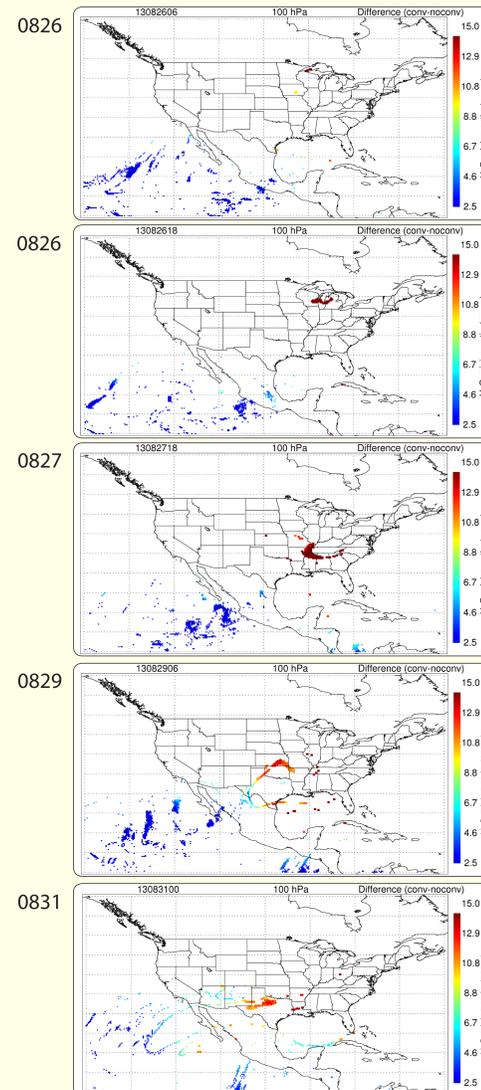
Case Study: MLS Water Vapor - Aug 26-27, 2013



MLS observed elevated water vapor near the Great Lakes at 100 mb and 83 mb from a storm on Aug 26, 2013. MLS was not positioned to see the elevated water vapor observed in situ on Aug 27, 2013.

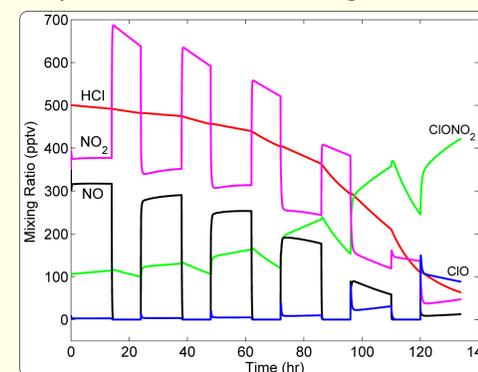
Case Study: Trajectory Model - Aug 26-31, 2013

Plots below are courtesy of Eric Jensen. P = 100 mb.



Anticyclonic flow pattern over the US is evident for elevated stratospheric water vapor from a deep convective storm near the Great Lakes on Aug 26, 2013. The elevated water vapor plume expands and experiences some dilution but stays relatively intact for ~5 days.

Case Study: Kinetic Model - late Aug 2013

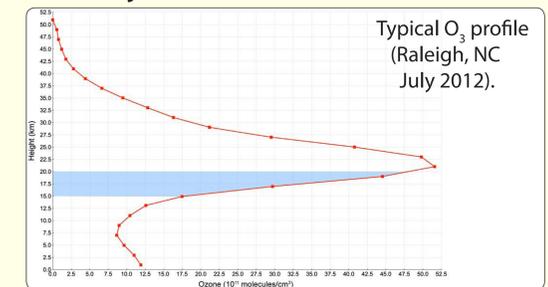


Calculated chemical impact of elevated water vapor for conditions representative of late August 2013. P = 90 mb, SA = 2 μm²/cm³, H₂O = 13 ppm, T = 203 K with radiative cooling of 0.05 K day⁻¹ ppm⁻¹ (Maycock et al. QJRM 2011; Dykema, in prep 2015), HCl interpolated from MLS, duration of elevated stratospheric water vapor = 5 days.

Key Questions:

- Will increases occur in the mixing ratio and altitude of injected water vapor?
- How long do plumes of elevated water vapor persist once injected?
- How much will stratospheric temperatures decrease due to climate change?
- How important is radiative cooling with elevated water vapor?
- Will geoengineering or volcanic eruption lead to elevated SA in the near term?
- How significantly will decreasing inorganic chlorine impact this mechanism?

Convective Injection, Ozone, and Altitude

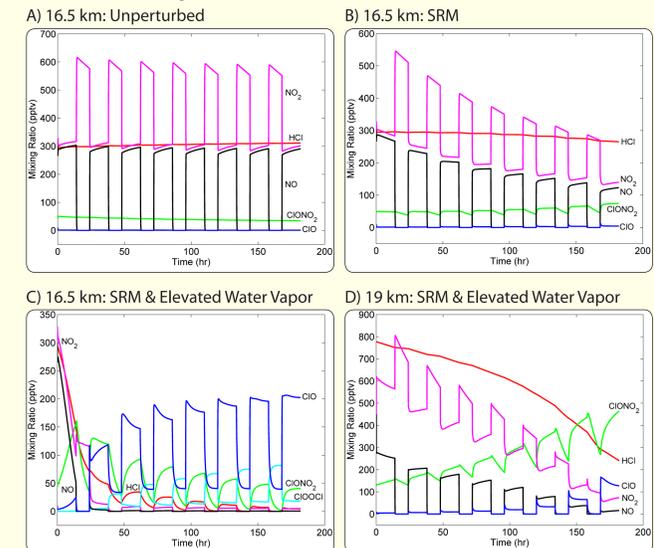


The potential for convectively injected water vapor to impact column ozone depends on the extent to which higher altitudes, where more ozone is present, become engaged.

| Variable | Altitude Dependence | Impact on % O3 Loss |
|-------------------------------|---------------------|---------------------|
| Temperature | Increases | Less |
| H ₂ O mixing ratio | Decreases | Less |
| H ₂ O duration | Increases | More |
| Inorganic chlorine | Increases | More |
| Reaction probability, γ | Decreases | Less |
| Aerosol surface area, SA | Decreases | Less |
| Nitric acid | Increases | Less |
| Ozone | Increases | Less |

Our sensitivity analysis (not shown) shows that the risk of ozone loss from elevated water vapor falls as altitude increases from 17 to 20 km, despite increasing inorganic chlorine. Percent ozone loss decreases with increasing altitude even if water is elevated.

Convective Injection and Aerosol Surface Area



Calculated chemical impact of elevated sulfate aerosol surface area from geoengineering or volcanic eruption and elevated water vapor. For the SRM cases, SA is increased from 2 to 20 μm²/cm³. For the elevated water vapor cases, H₂O is increased from 5 to 12 ppm. Starting T = 202 K at 16.5 km and 206 K at 19 km with radiative cooling. HCl, HNO₃, and O₃ are from MLS.

Summary

The highest mixing ratios of stratospheric water vapor observed at the highest latitudes globally are over the United States in summer. Aided by the anticyclonic flow pattern in the lower stratosphere resulting from the NAM, convectively injected high water plumes can stay relatively intact for over a week. A modeled sensitivity analysis shows that the potential impact on inorganic chlorine and ozone from elevated water decreases as altitude increases above 90 mb even with convective injection reaching higher altitudes. The combination of elevated water vapor with elevated sulfate aerosol surface area from geoengineering or volcanic eruption has significantly greater potential to impact inorganic chlorine and ozone than changing either variable alone. Any chlorine activation and ozone loss initiated by convective injection of water is highly dependent upon the variables listed in the table above with temperature being most important.