Quantifying and confirming our understanding of processes controlling stratospheric ozone: Insights from UARS and Aura MLS

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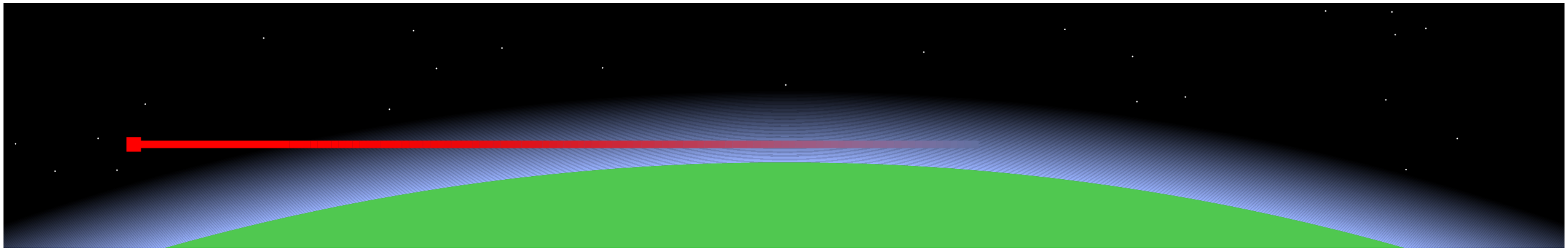
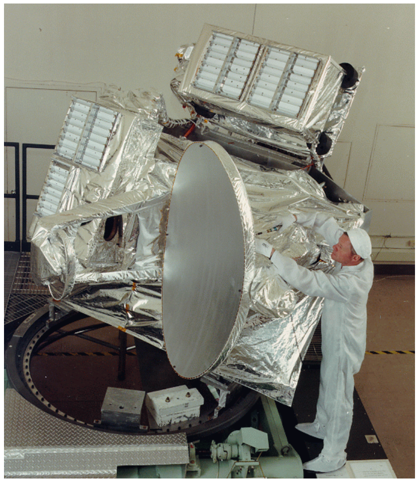
The microwave limb sounding technique

- Measurement of millimeter- and submillimeter-wavelength thermal emission from rotational lines of atmospheric molecules as the instrument field of view is vertically scanned through the atmospheric limb
- Many atmospheric gases, including key species in stratospheric ozone chemistry as well as several markers of pollution, have spectral lines in the microwave region
- Advantages over nadir sounding: observing the atmosphere "edge on" provides much better vertical resolution; no need to characterize surface variations (e.g., emissivity); longer ray path through the atmosphere gives a stronger signal for tenuous trace gases
- Advantages over UV, visible, and IR instruments: long-wavelength measurements are much less sensitive to thin clouds or volcanic aerosol
- Advantages over scattering and occultation: continuous measurements day and night
- Disadvantages: poorer horizontal resolution than achievable with nadir sounding; continuum absorption limits microwave limb sounder observations to the atmosphere above ~400 hPa (~8 km); large antenna needed for best vertical resolution



MLS on the Upper Atmosphere Research Satellite (UARS): 1991–2001



- The first Microwave Limb Sounder (MLS) was one of 10 instruments deployed from the Space Shuttle in 1991 as part of NASA's UARS mission designed to enable comprehensive study of the chemistry, dynamics, and radiation of Earth's atmosphere
- UARS MLS was the first space-borne limb-viewing instrument making measurements in the microwave
- With radiometers at 63, 183, and 205 GHz, its primary measurement objectives were stratospheric ozone, water vapor, temperature, and ClO (chlorine monoxide), the primary agent of ozone destruction in the stratosphere
- UARS MLS operated for ~10 years (with observations becoming increasingly sparse after ~1995)

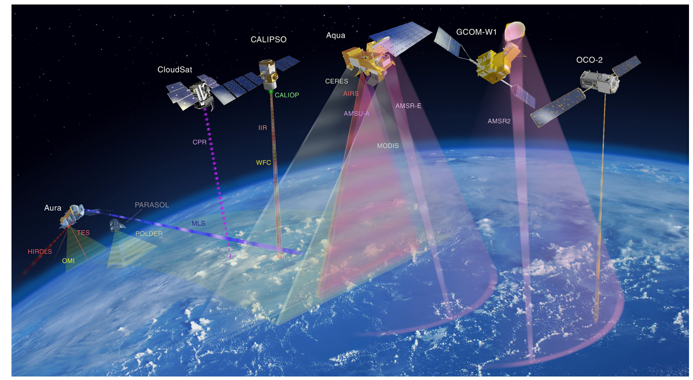
UARS MLS discovery: Reactive chlorine fills the Antarctic and Arctic vortices

- Localized measurements from ground-based and aircraft instruments had indicated that:
  - Heterogeneous chemical reactions on the surfaces of polar stratospheric cloud (PSC) particles – which form when lower stratospheric temperatures drop low enough – convert inactive reservoir chlorine (e.g., HCl, $ClONO_2$) into ozone-destroying forms
  - The resulting enhancements in ClO are highly correlated with ozone destruction
  - Such reactions take place in both polar regions
- The first maps of stratospheric ClO, made a few days after deployment of UARS in September 1991, showed that reactive chlorine pervades the Antarctic vortex
- Arctic ClO enhancement can be comparable, in both magnitude and areal extent, to that in the Antarctic
- Active chlorine is contained within the polar vortices
- UARS MLS revealed the full 3D distribution of seasonal chlorine activation and its correlation with lower stratospheric ozone depletion in both polar regions, and allowed quantification of chemical loss in the Arctic

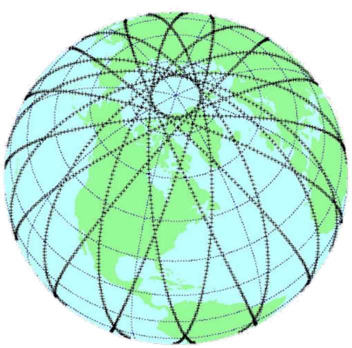
Waters et al., Nature, 1993; see also Manney et al., Nature, 1994; Santee et al., Science, 1995

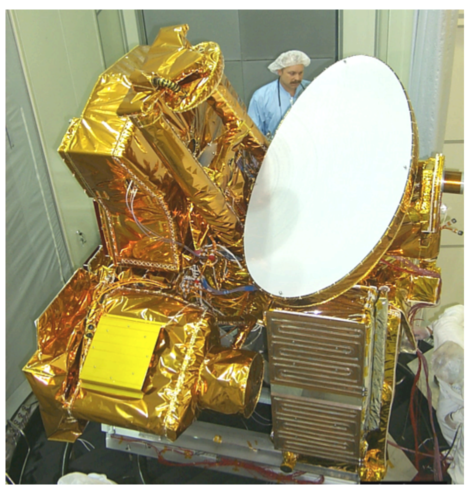
MLS on the Earth Observing System (EOS) Aura satellite: 2004–present

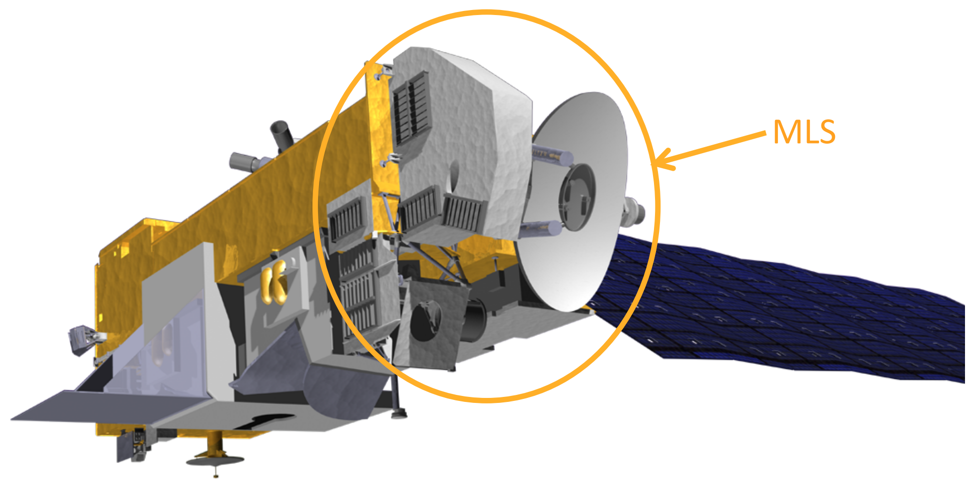
- Aura is a NASA satellite mission dedicated to addressing these overarching questions:
  - Is the stratospheric ozone layer changing as expected?
  - What are the processes controlling air quality?
  - How is Earth's climate changing?



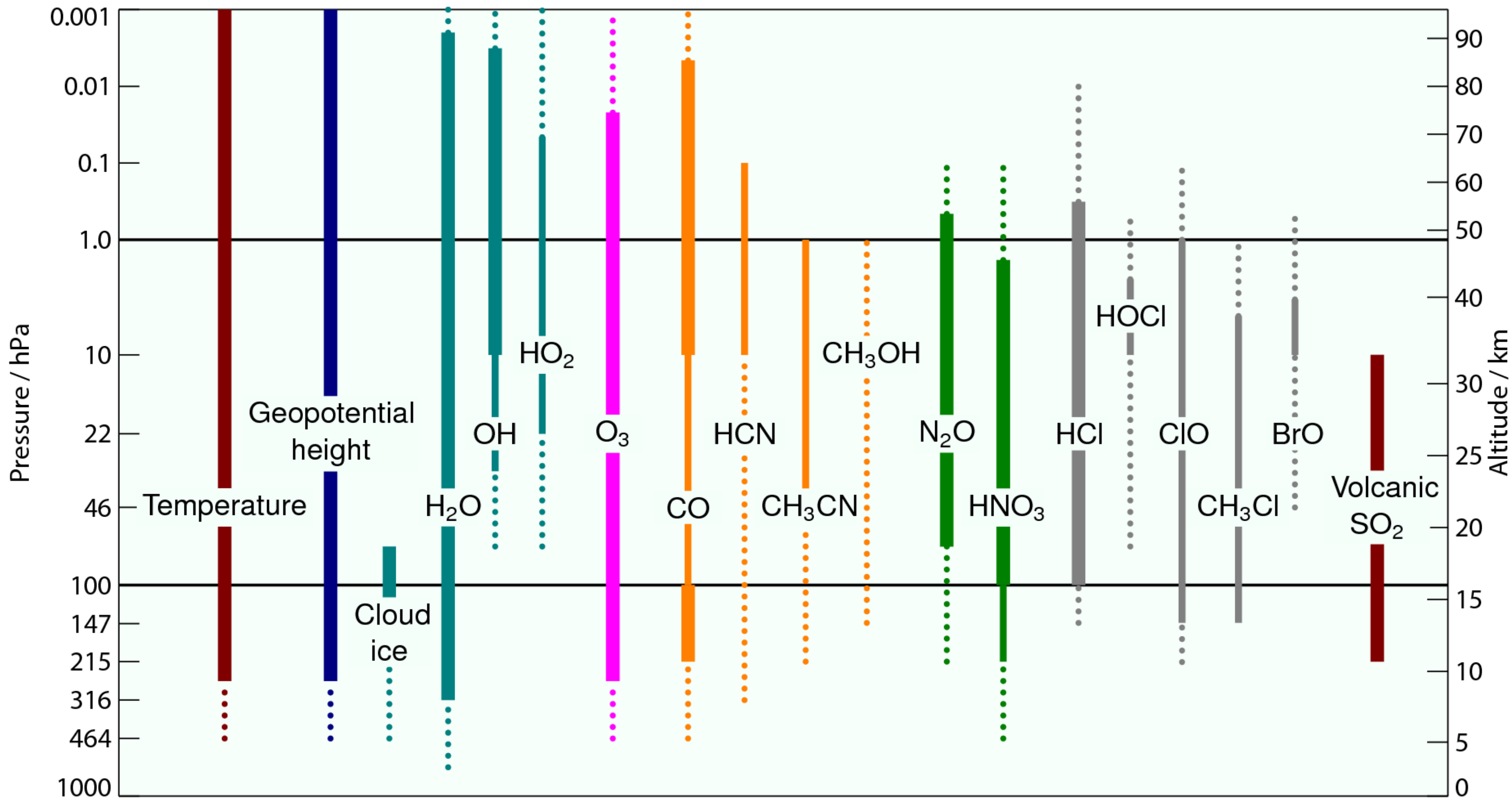
- Launched in July 2004, Aura flies in formation in the "A-Train" constellation of satellites
- With continuous global coverage from 82°S to 82°N on every orbit, Aura MLS obtains 3500 vertical profiles daily
- Aura MLS has radiometers in 5 broad spectral bands centered near 118, 190, 240, 640, and 2500 GHz (wavelengths from 2.5 to 0.1 mm)





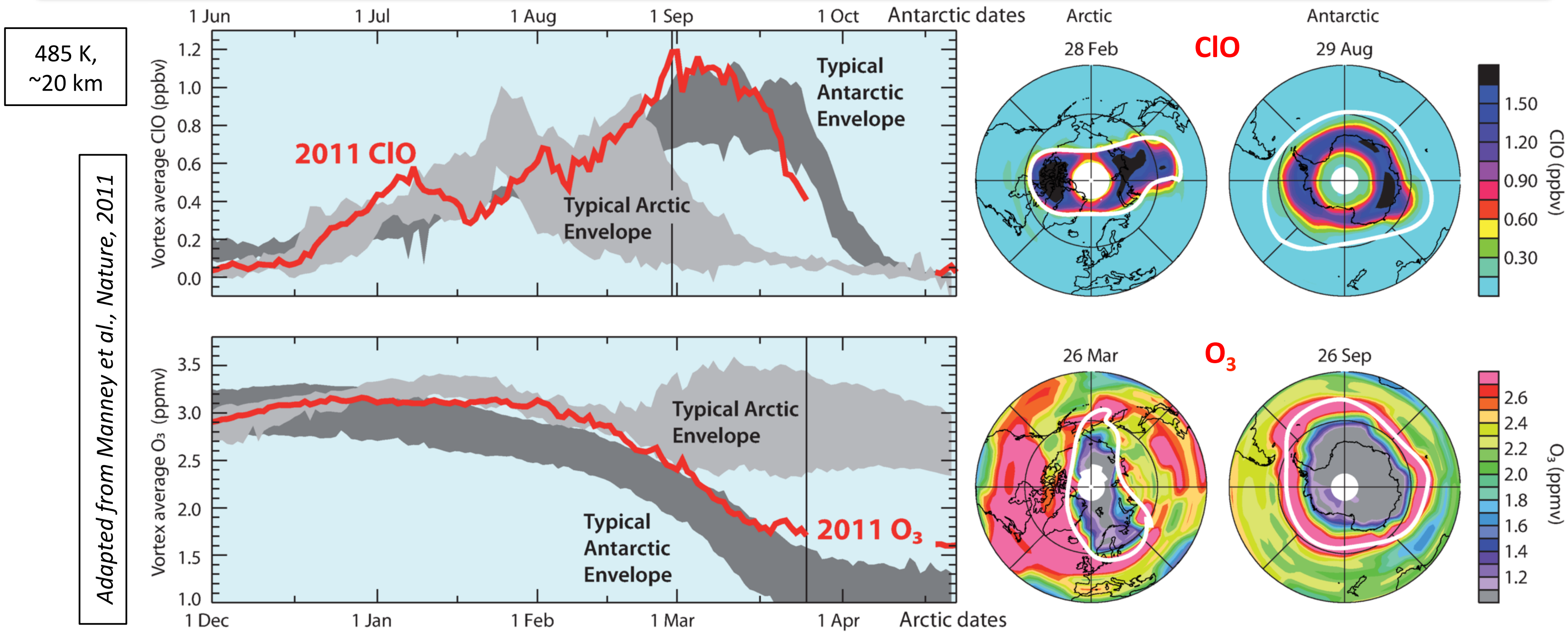


Aura MLS measurement products (version 4)



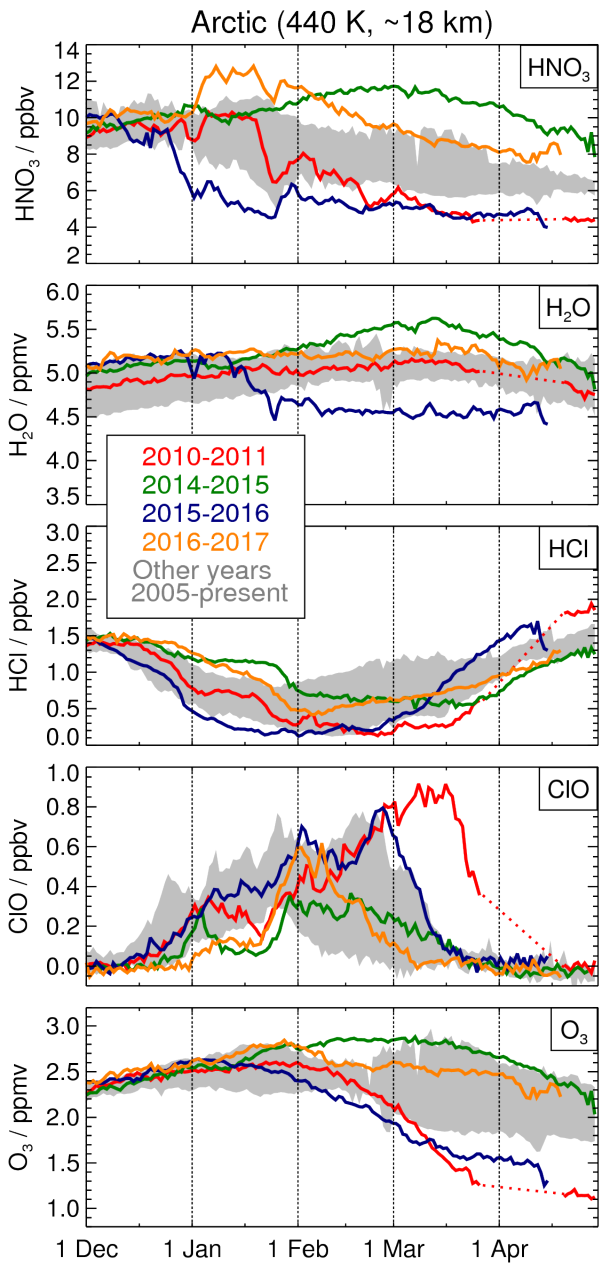
Aura MLS makes several measurements of relevance for stratospheric ozone chemistry, including both reservoir (HCl) and reactive (ClO) chlorine, the primary building blocks of PSCs (HNO_3 and H_2O), and a tracer of stratospheric air motions (N_2O), as well as ozone itself

Unprecedented Arctic ozone loss in 2011



- Minimum temperatures were below the chlorine activation threshold for a longer time and over a deeper vertical region in 2010/2011 than in any other Arctic winter on record
- ClO enhancement in 2011 was much more intense, extensive, and prolonged than in any other cold Arctic winter observed by MLS, with peak ClO abundances starting to decline rapidly only ~1 week earlier than typical in the equivalent season in the Antarctic
- An unprecedented degree of chemical ozone loss, coupled with atypically weak transport into the lower stratospheric polar vortex, led to exceptionally low springtime ozone

Interannual variability confounds detection of polar ozone recovery: Arctic

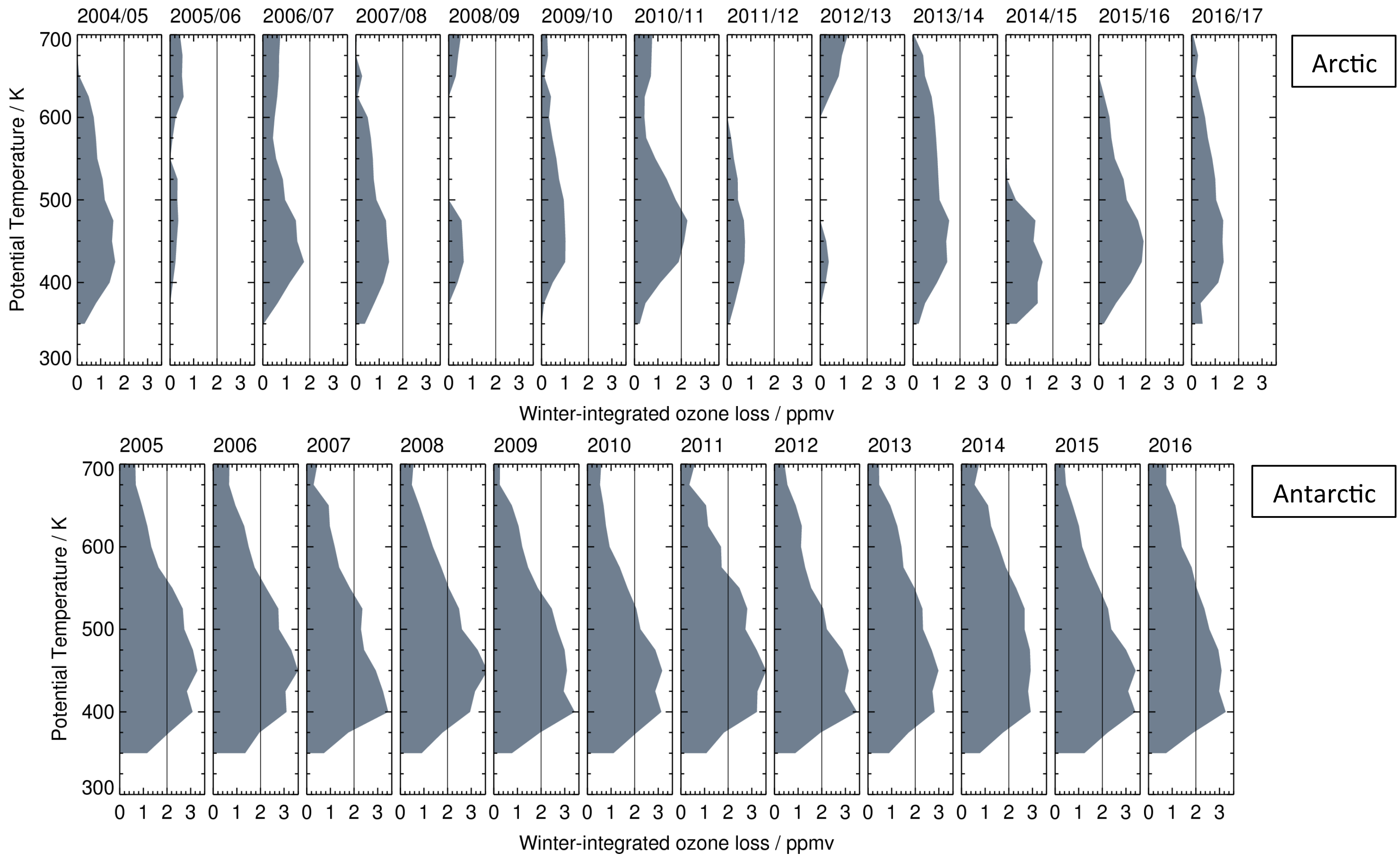


- The Arctic winters observed by Aura have been extremely dynamically diverse, including several when sudden stratospheric warmings curtailed chemical ozone loss early
- For example, conditions in the 2014/2015 winter/spring (green lines) were not conducive to significant ozone loss
- It was again extraordinarily cold in 2015/2016 (blue lines):
  - Unmatched (for the Arctic) denitrification / dehydration
  - Strong early winter chlorine activation
  - Ozone values comparable to or slightly below those in 2010/2011 (red lines) through much of the winter
  - But a major final warming in early March 2016 halted chemical processing about a month earlier than in 2011, keeping ozone destruction below record levels
- Such large meteorologically driven variability makes detection of significant trends in ozone (and other species) a particular challenge in the Arctic

Adapted from Manney et al., ACP, 2015; Manney & Lawrence, ACP, 2016

Quantifying polar stratospheric ozone loss: A Match-based approach

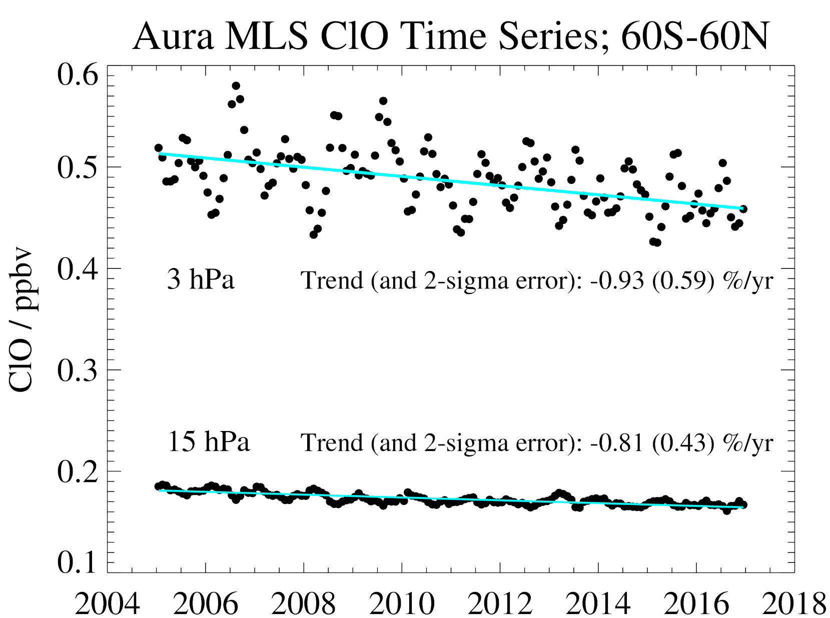
- The Lagrangian "Match" technique accounts for transport effects by measuring changes in ozone between two observations of the same air mass, thus quantifying the chemical loss
- Profiles of cumulative chemical ozone loss show strong interannual variability in the Arctic; in the Antarctic, loss is much larger, deeper, and more uniform from year to year

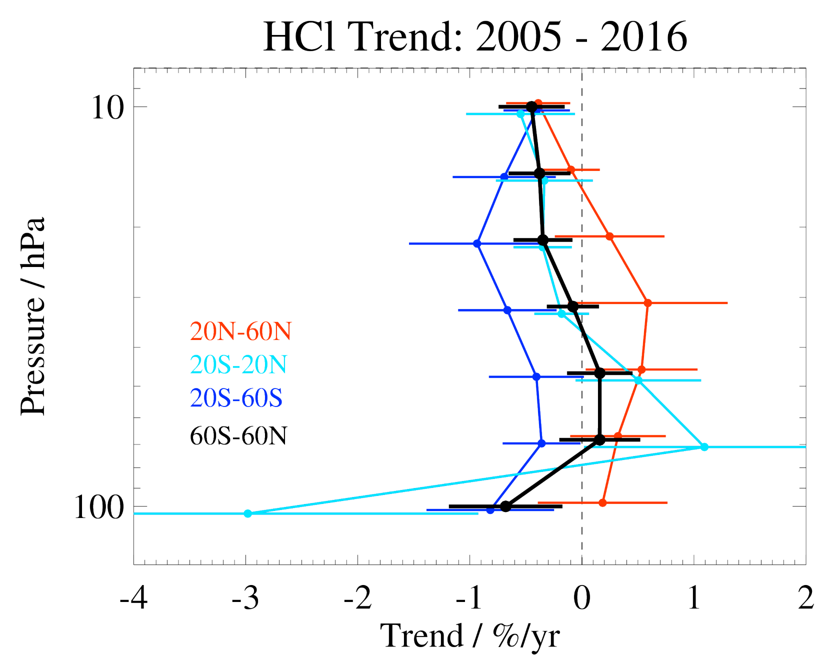


Adapted from Livesey et al., ACP, 2015

Decline in stratospheric chlorine species

- Tracking the evolution of stratospheric chlorine using the 13-year (to date) record of Aura MLS HCl and ClO measurements is an area of active research
- Global-mean ClO data indicate statistically significant reductions of ~0.8–0.9 %/yr in the upper stratosphere over the Aura timeframe, consistent with the continuing decline in ozone-depleting substances controlled under the Montreal Protocol
- However, MLS HCl in the lower stratosphere shows hemispheric asymmetry, with decreasing trends in the Southern Hemisphere but mostly positive or non-significant trends in the Northern Hemisphere and the tropics; this variability (still under investigation) likely reflects the influence of changes in the atmospheric circulation





Figures courtesy L. Froidevaux, 2017

Summary

- The Microwave Limb Sounder (MLS) instruments on NASA's UARS and Aura missions have provided an extensive suite of trace gas measurements enabling quantification of the chemical and dynamical processes controlling the distribution of stratospheric ozone
- The occurrence in recent winters, especially in the Arctic, of different combinations of extreme meteorological conditions, and their critical influence on chemical processing, underscores the lower stratosphere's sensitivity to a spectrum of dynamical variability
- Such sensitivity argues for the importance of continuing comprehensive daily global composition measurements to enable diagnosis of unexpected variations
- However, the future of space-based microwave limb sounding is highly uncertain – no agency or country currently has any concrete plans for continuing such measurements once Aura MLS (now well past its 5-yr design lifetime) is no longer operational