



Atmospheric HCI Variations from MLS and studies of data continuity

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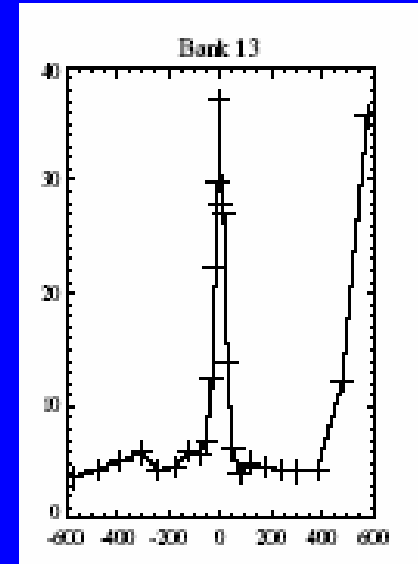
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- **The Microwave Limb Sounder (MLS) measures**

- HCl from thermal emission near 626 GHz**

- Radiance residuals are typically < 1%
 - good radiance fits (closure) in V1.51 data
- Single profile precision is between 5 and 15%
 - 0.3 - 0.4 ppbv in upper stratosphere/lower mesosphere
(focus of this presentation)
- Vertical resolution is ~ 3 km (lower strat.) to 6 km (upper strat.)
- Recommended vertical range: 100 to 0.2 hPa
 - but sensitivity extends to upper mesosphere
- Early validation results in IEEE Aura Special Issue [Froidevaux et al., in press]
 - Besides satellite/satellite comparisons, have shown that MLS HCl is consistent overall with balloon HCl data [1st validation flight, Ft. Sumner, Sep. 04]
(few statistics for clear view of biases; analyses still in progress for 2nd flight)



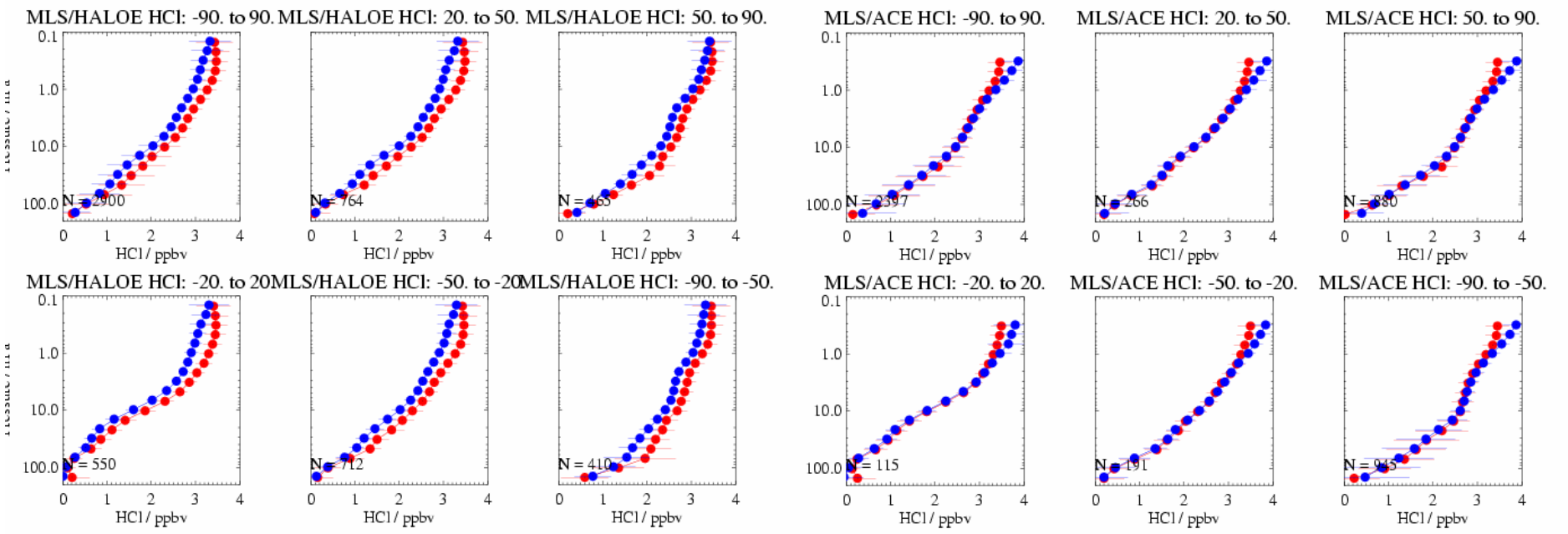
HCl emission line
R4 radiometer
Band 13 (626 GHz)



Compare averages of > 2000 coincident profiles (147 hPa to ~ 0.1 hPa)
 find closest MLS profiles in range $\Delta\text{Lat} = \pm 2$ deg., $\Delta\text{Lon} = \pm 8$ deg., $\Delta\text{time} = \pm 12$ hr (same day).

MLS (V1.5) vs HALOE (V19)
 Aug. 04 – Jul. 05

MLS (V1.5) vs ACE (V2.2)
 Aug. 04 – Aug. 05



- **MLS HCl > HALOE HCl by ~ 5 - 15%; MLS → evidence of HCl decrease up high.**
- **MLS HCl is generally within 5% of ACE HCl; ACE looking into diff. at topmost levels**

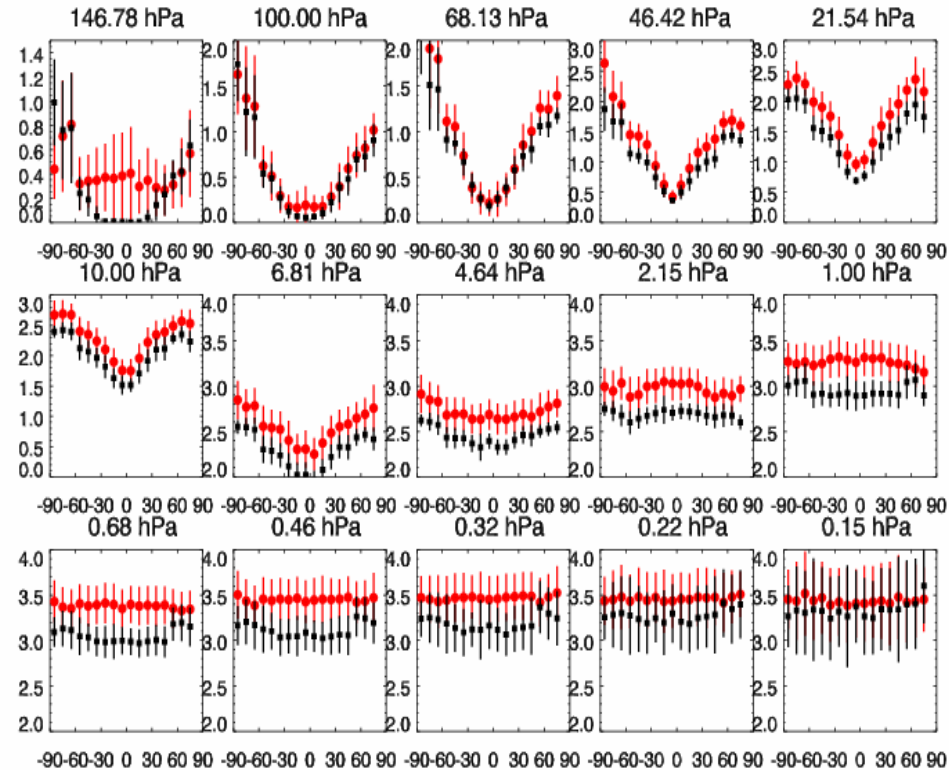


Variation of HCI (avg. coincident profiles for 1 yr) versus latitude

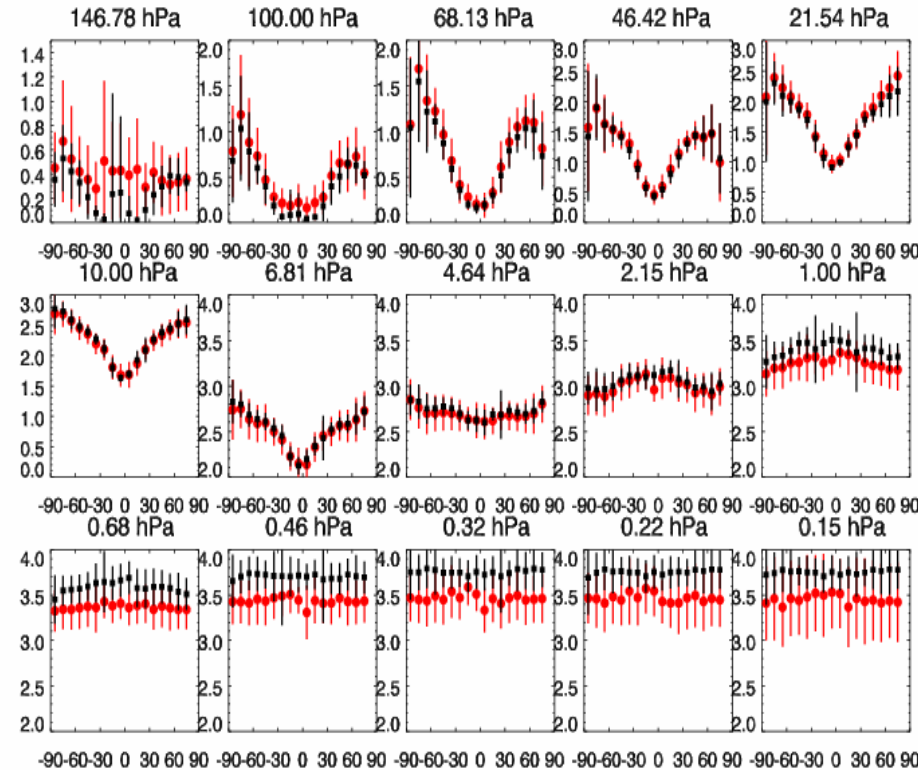
MLS vs HALOE

MLS vs ACE

MLS and HALOE Avg. Coincidences vs Latitude: HCI



MLS and ACE Avg. Coincidences vs Latitude: HCI



- Good tracking of latitudinal variations: stable to a few % down to 50 hPa
- MLS values at 147 hPa appear too large, mainly equatorward of ~ 40° latitude
- Diffs. & σ_{dif} also fairly stable versus time (month) over past year [not shown here]

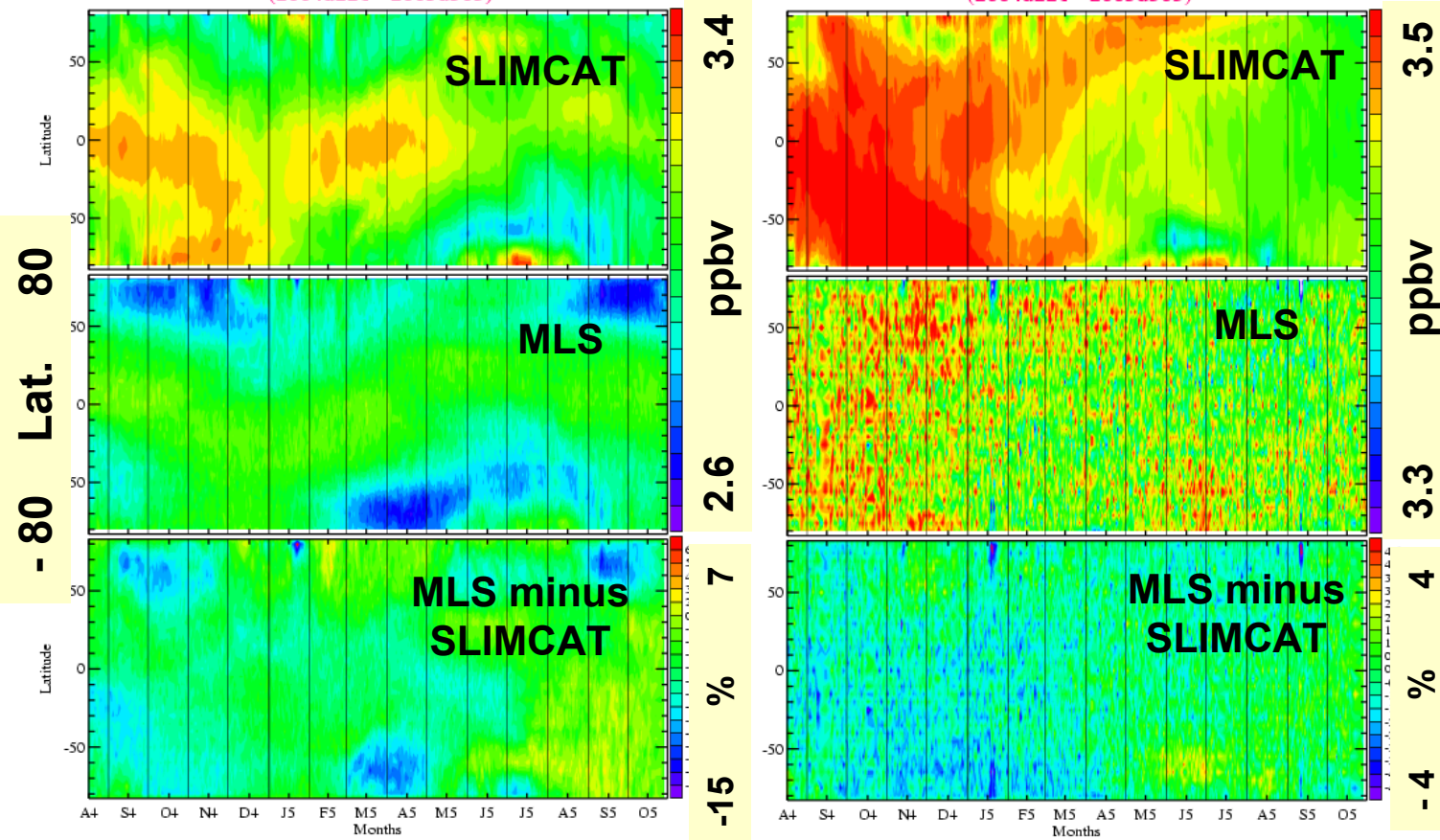


MLS HCI vs SLIMCAT 3-D CTM: upper stratosphere / lower mesosphere

Model was run daily for past year; files have sampling 'a la MLS'

2 hPa
HCI Zonal Mean Time Series (P= 2.15 hPa)
SLIMCAT (top), MLS (center), MLS-SLIMCAT (bottom)
(2004d221 - 2005d303)

0.5 hPa
HCI Zonal Mean Time Series (P= 0.46 hPa)
SLIMCAT (top), MLS (center), MLS-SLIMCAT (bottom)
(2004d221 - 2005d303)



Overall, similar variations are observed in SLIMCAT and MLS fields (+ small offset, other diffs.)

2 hPa: Seasonal and semi-annual patterns exist

0.5 hPa: Slow decrease over time is noticeable

Aug. 04

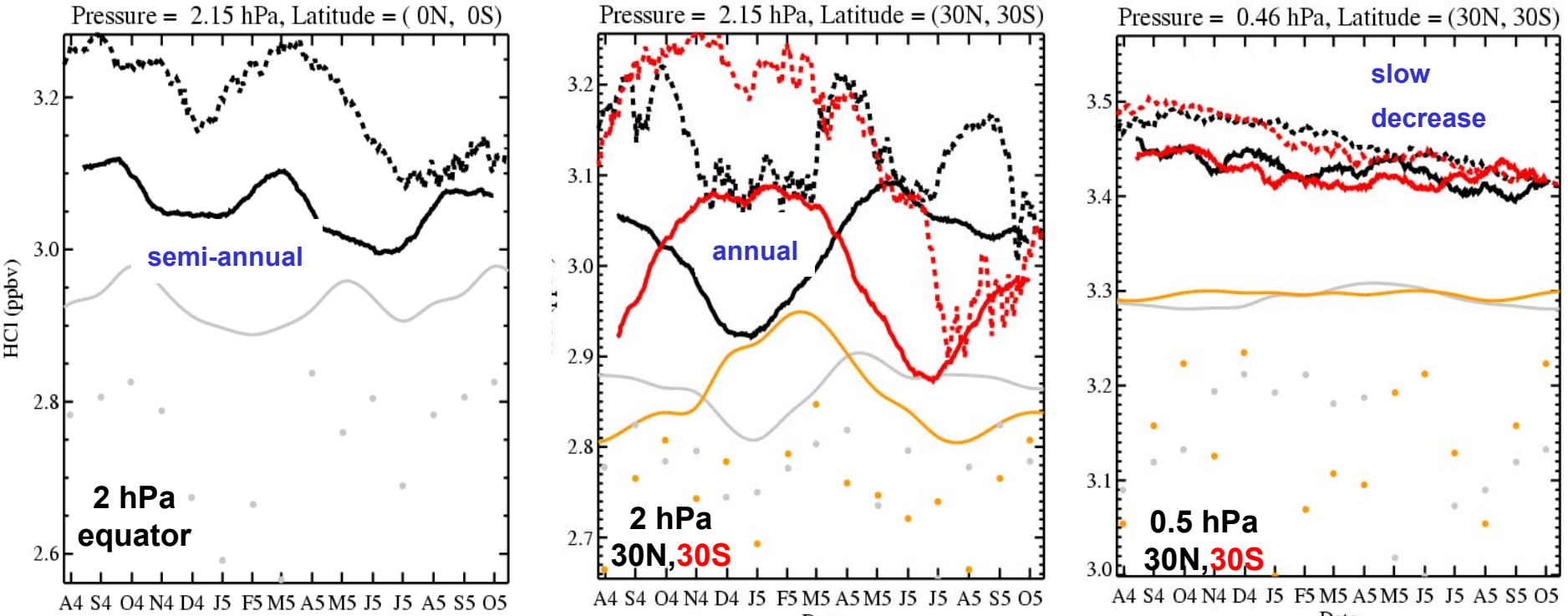
Oct. 05

Aug. 04

Oct. 05



MLS HCl vs SLIMCAT 3-D CTM: upper stratosphere

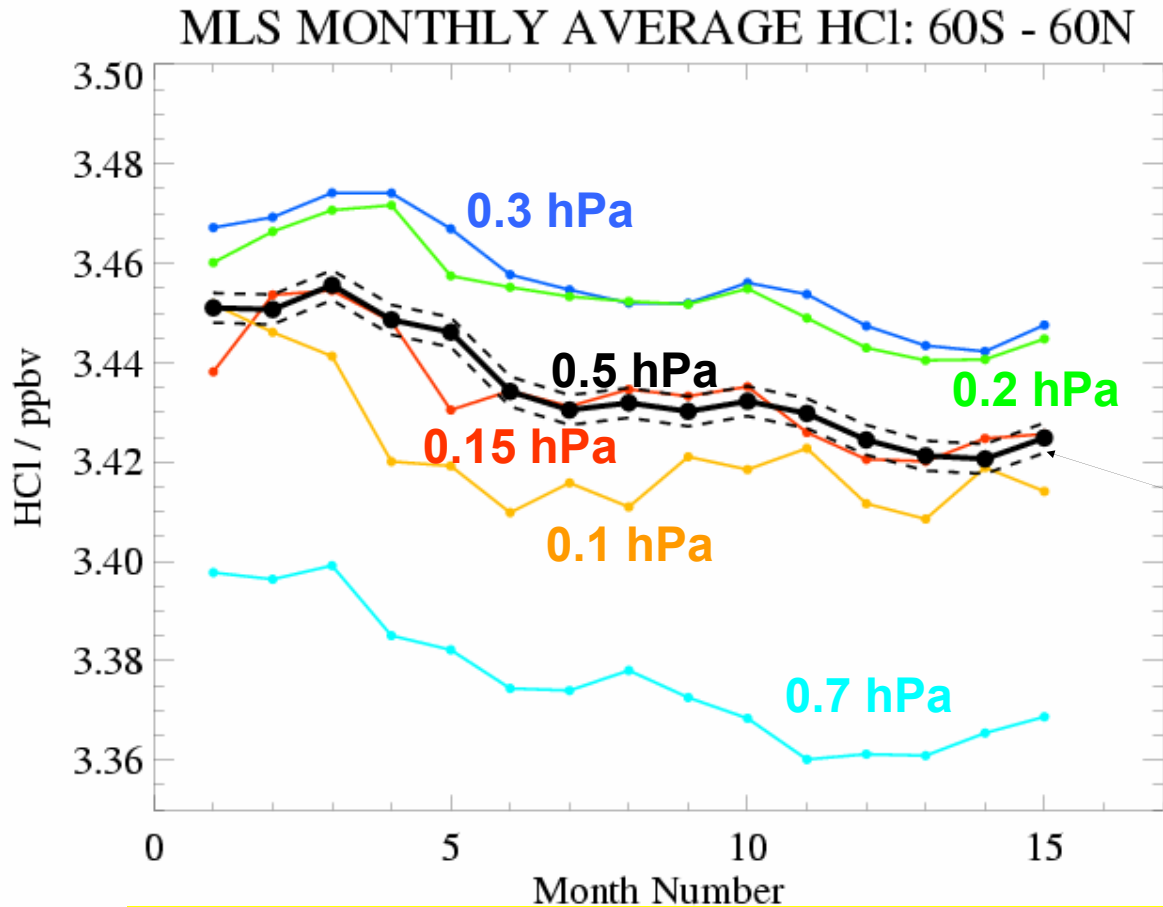


Legend: 30N: black & grey 30S: red & orange SLIMCAT: dashed
 MLS: solid (time series smoothed with 30 day window); 1 σ var. within month ~ 0.2-0.3 ppbv
 HALOE Climatology (from 5 past years): grey & orange dots
 A priori MOZART climatology [D. Kinnison, NCAR]: grey & orange curves

A priori (climatology) has negligible influence (< 0.5 %) on retrievals at 0.5 hPa

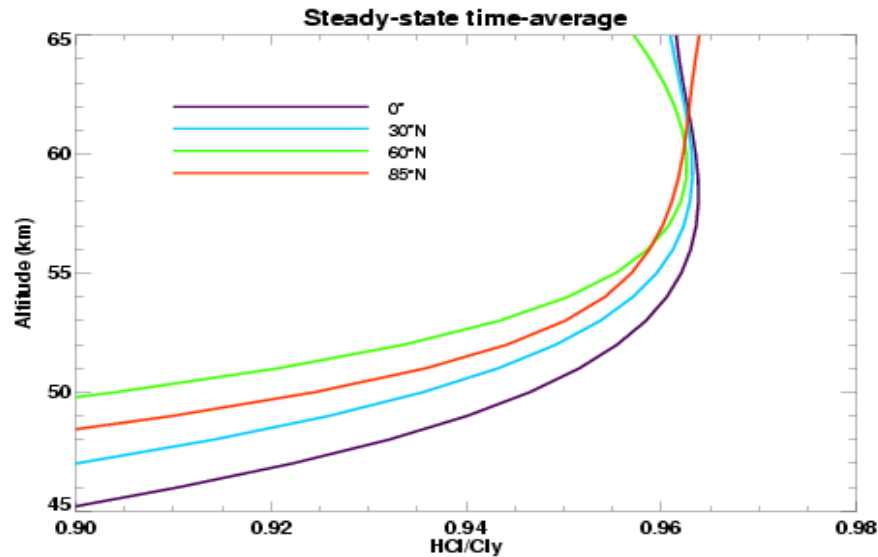


HCl decrease in upper stratosphere and mesosphere



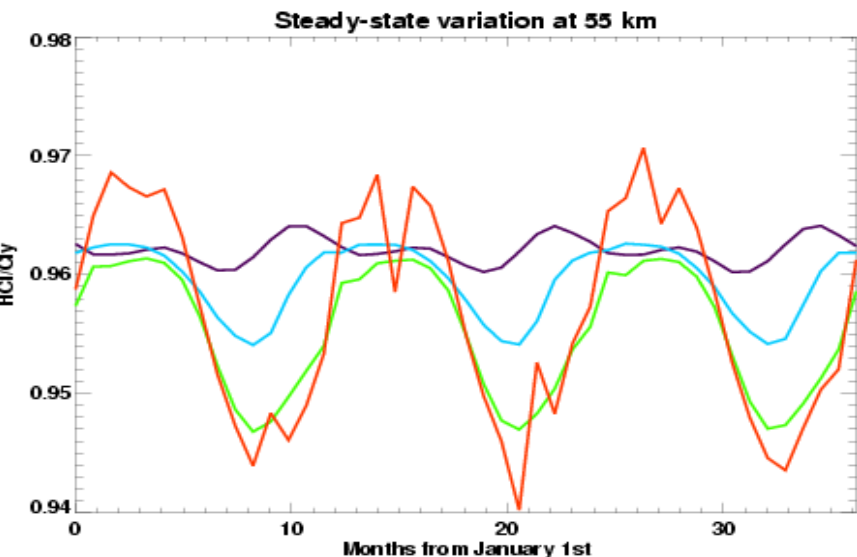
Aug. 04 Oct. 05

- Slow and fairly steady decrease in global HCl Aug. 04 to Oct. 05
→ decrease = 1.0% or ~ '20 σ ', given that MLS prec. (2σ) < 3 pptv
- Expect ~ 1% decrease from surface chlorine (source gas decreases).
- Results are very similar for 30S - 30N or for 90S - 90N.
- In early 2005:
0.5 hPa HCl = 3.43 ppbv
→ Cly = 3.61 ppbv (close to ACE value)
Error bars on later slide.



- To compare HCl and Total Chlorine (Cly) in upper stratosphere / lower mesosphere, need to correct for ratio of HCl / Cly.

> Model values for HCl/Cly at left come from the NCAR SOCRATES 2-D Model [thanks to the team that made this available].

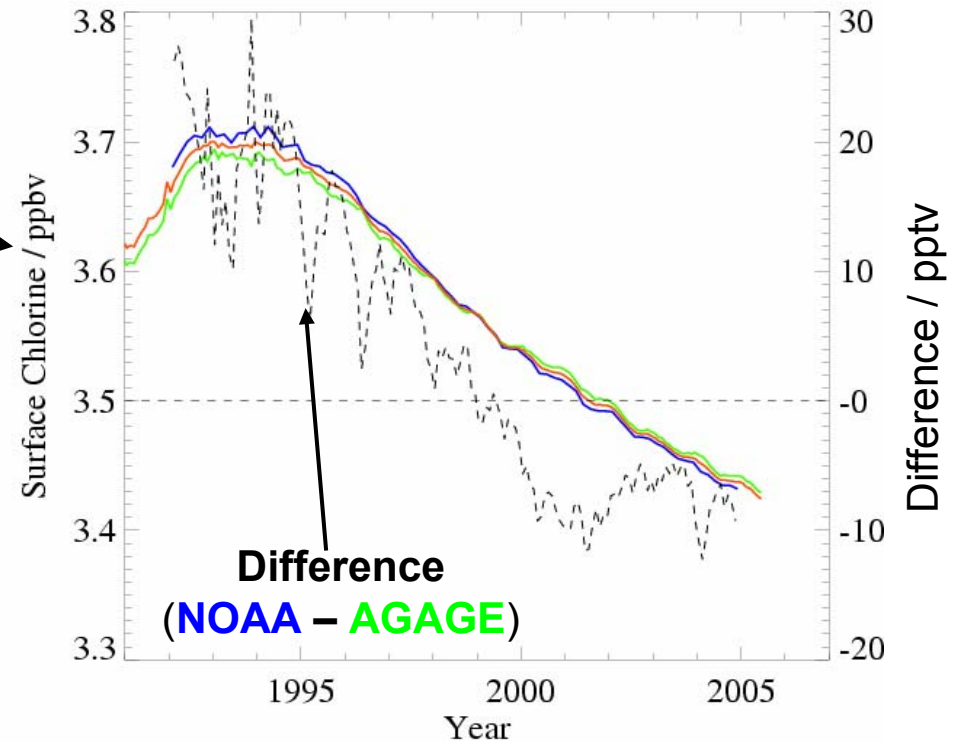
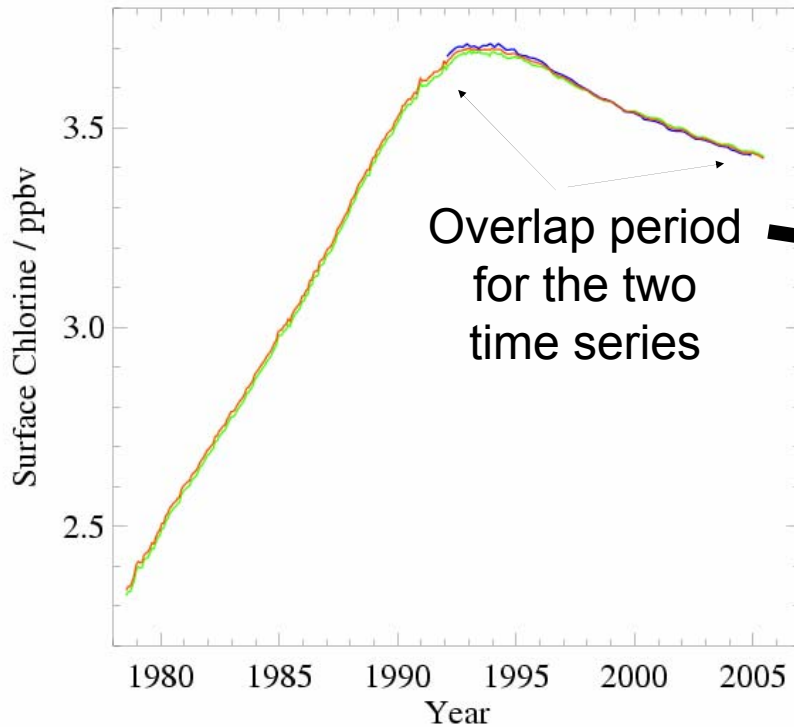


→ for HCl near 55 km (0.5 hPa), use HCl/Cly = 0.95 (uncertainty ~ 0.01).

> Model values in SH very similar to these.



Surface Chlorine Time Series



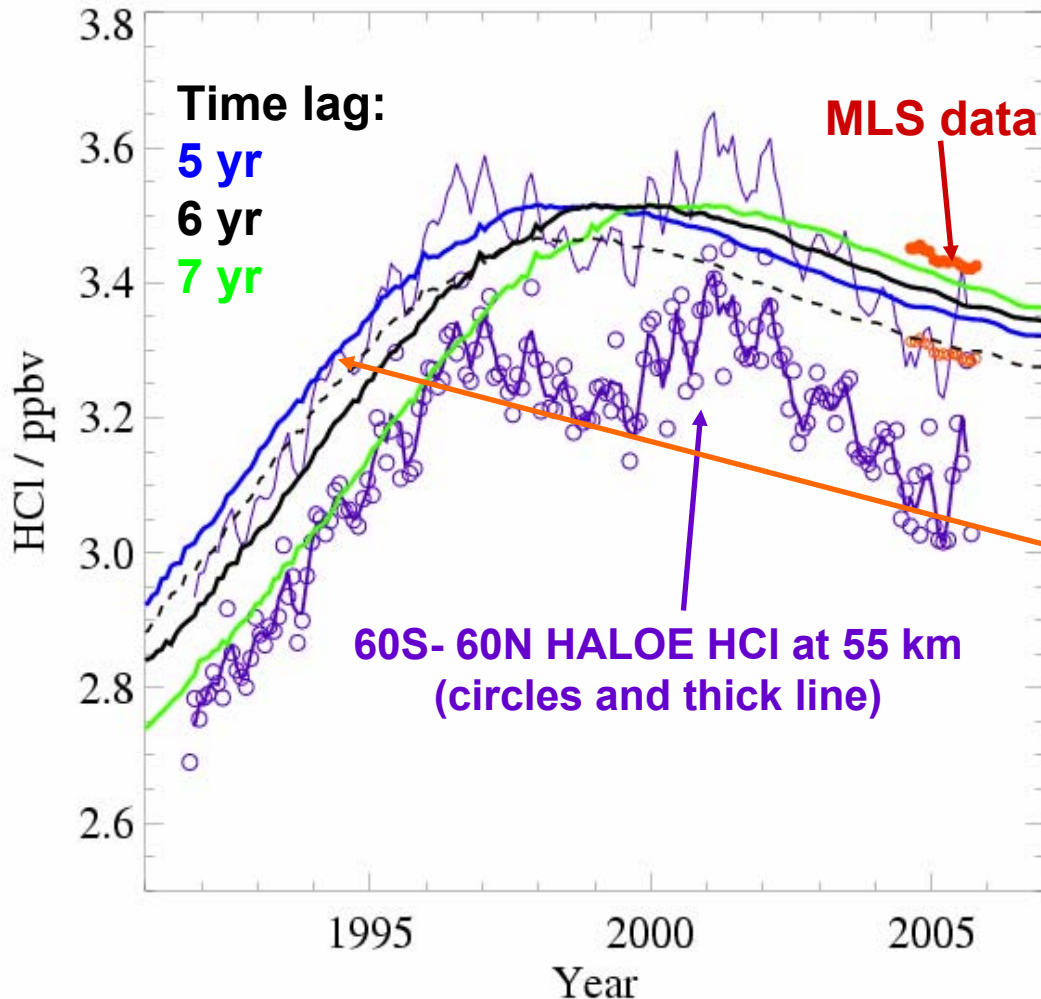
Mean surface chlorine curve is made from average of **AGAGE** and **NOAA** values

- Difference between the two estimates varies between - 20 pptv and 30 pptv; this is the type of uncertainty expected in total surface chlorine, i.e. ~ 1%.

- Surface (organic) chlorine peaks near 3.7 ppbv → max. value for stratosphere.



Total Chlorine: surface versus satellite data

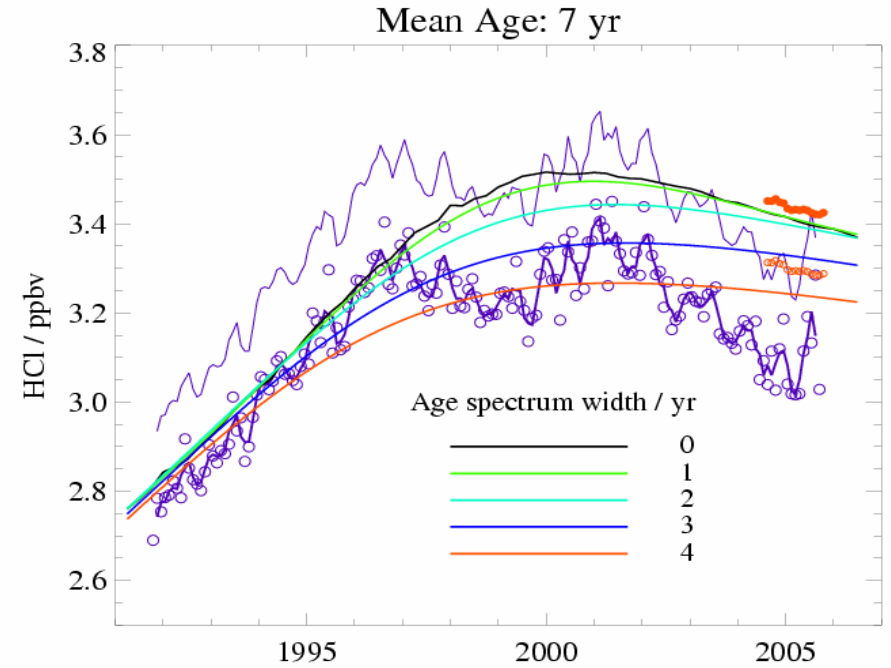
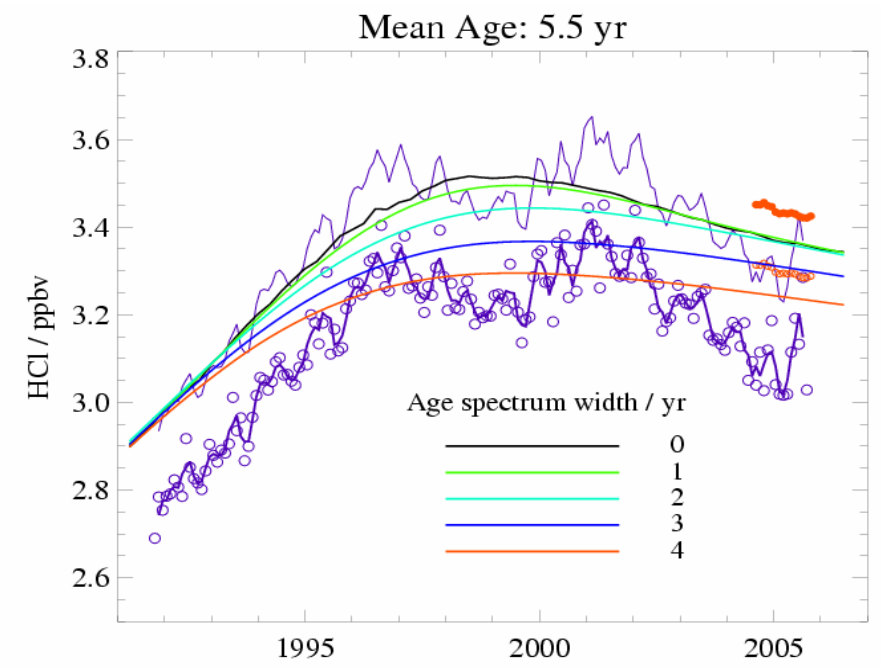


- The MLS HCl decrease matches expected slope from simple time-lagged surface chlorine curves (shifted by 5, 6, 7 years and translated to HCl - factor of 0.95).
- 'Adjusted' dataset possibility: add 7 % to HALOE, (thin purple curve) and subtract 4% from MLS (open red circles); consistent with quoted error bars – but not uniquely. **This would also match ATMOS data better [1994 Cly data – Zander et al.]**
- Can fit 'adjusted dataset' with time shift of ~ 5 yr; black dashes assume ~ 50 pptv Cl does not get to 55 km.
- Some HALOE variations are not explained by these curves, but also, MLS (& ACE) data 'on the high side'.



Mean age and age spectra considerations

Concepts of mean age and age spectra (from mixing & transport of parcels with different origins and 'ages') will play a role in changing stratospheric chlorine curves from a simple time lag of surface values [e.g., Waugh et al.].



- MLS 04-05 do not constrain age/width very much: better if mean age ~ 7 yr & width not > 3.
- Early data (HALOE) should 'fix' mean age better, but also sensitive to total (Cly).
- Can fit 'adjusted dataset' (thin purple) best with ~ 5.5 yr mean age and width of 0 to 3.



Main error sources for MLS retrievals	Impact on HCl near 0.5 hPa (‘2 sigma’ estimates)
Radiance noise	<ul style="list-style-type: none"> - Single Profiles: 20% (0.6 - 0.8 ppbv) - Monthly Global Avgs. : < 0.1% (2 - 3 pptv)
<i>a priori</i> influence	0 to - 0.5%
Calibration (radiometric & spectral)	1.5 %
HCl spectroscopy	3 %
FOV, pointing, Ptan, T	< 2 to 3 %
Retrieval closure accuracy	1 %
Total absolute error estimate	rss: 4 to 5 %



Brief Summary

- MLS HCl abundance variations (vs latitude and time) compare well with coincident HALOE and ACE profiles after 1 year of data.
 - but some offsets exist (MLS > HALOE; MLS ~ ACE, except at highest altitudes).
- MLS HCl variations with latitude and season are well captured, overall, by the SLIMCAT 3-D CTM results [upper & lower strat.] → some ‘sanity check’.
- MLS HCl t-series → good agreement w/ expected source gas (CFC) decreases (see also ground-based column data – HCl, ClONO₂; *Rinsland, Mahieu*)
 - (a) slope/change, 2004 to 2005 [MLS first 12 months of data → ~ 0.8 % decrease]
 - (b) absolute values - within ~ 5 % MLS error bar + other errors, e.g. total chlorine, HCl/Cl_y, but MLS seems a bit on the ‘high side’ (for time lag < 7 yr).
- Difficult to completely reconcile the HALOE and MLS time series (abs. & rel.).
 - > Some HALOE HCl time dependence (low frequency variability) seems difficult to explain by expected changes shown here, and by MLS slow decrease in HCl; sampling could play some role in this variability (will be looked at).
 - > Mean age of ~ 5.5 years and age spectrum width < 3 yr give best overall fit.