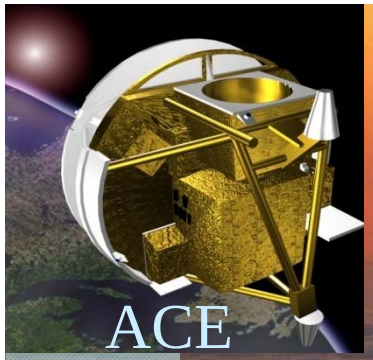


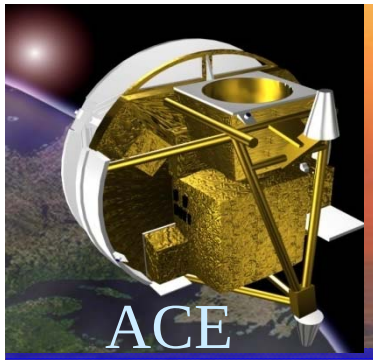
Retrievals for the Atmospheric Chemistry Experiment (ACE) Satellite Mission

Chris Boone, Kaley Walker, & Peter Bernath

7th Atmospheric Limb Conference

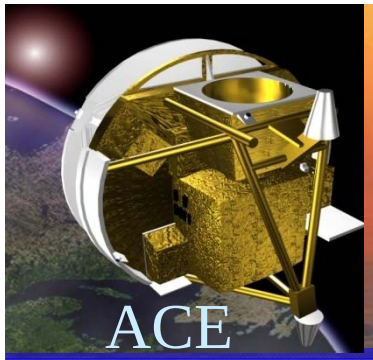
June, 2013

University of Waterloo



Introduction

- As per Kaley's talk
- The Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS)
- Maximum optical path difference ± 25 cm
- Wavenumber range 750 – 4400 cm^{-1}
- Solar occultation. SNR ~ 300:1.
- Collects measurements every 2 s, samples the atmosphere at 2-6 km intervals.



ACE-FTS Processing Status

- Versions 2.2 and 3.0 have stopped processing. Results from October 2010 onward in both versions unreliable. Problems with P and T info from the CMC.
- Version 3.5 in progress. Priori to October 2010, results will be the same as version 3.0, except for HCFC-22, CO above 60 km, C_2H_6 near 20 km, and N_2O for a number of occultations with low H_2O (where the N_2O retrieval failed to converge).
- Version 2.5 also processing. Prior to October 2010, results will be the same as version 2.2, except HDO, N_2O_5, and O_3 updates will be incorporated into the main output files.

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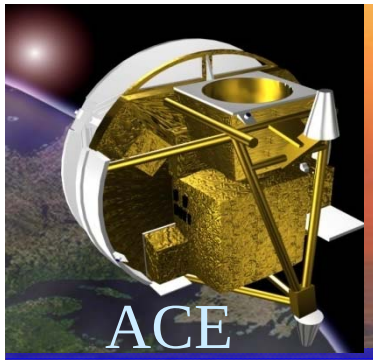

CMC Data Problems

- Two sets of a priori pressure and temperature data obtained from the CMC: one set from a global model and one from a regional model.
- We have always used the results of the regional model, assuming a slight accuracy advantage.
- In the p/T retrievals, pressure and temperature below 12 km in v2.2 or below 15 km in v3.0 are fixed to the CMC data.
- October 2010, something went wrong. Problem interpolating profiles to measurement locations rather than a problem with the model.

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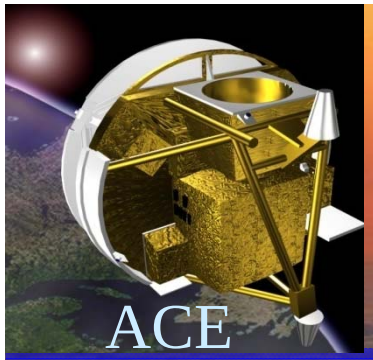
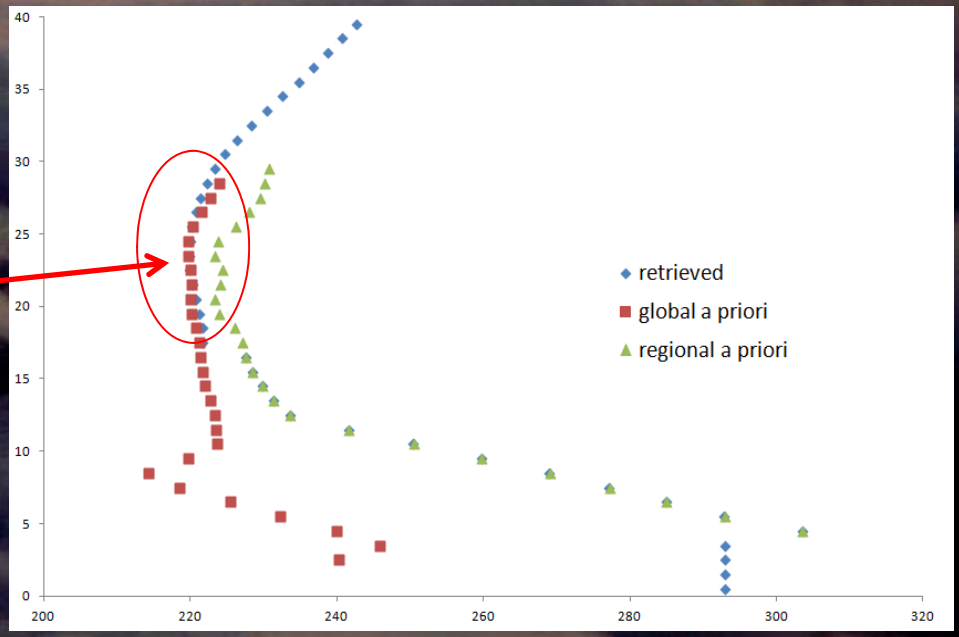


Example (sr40906)

- Regional results are unphysical. In a polar occultation, > 30 °C at 4.5 km.
- Versions 2.5 and 3.5 use pressure and temperature from the global model.

Retrieved v3.0 temperature profile agrees well with the CMC global-model data, even though those data were not used in the analysis, which is encouraging.



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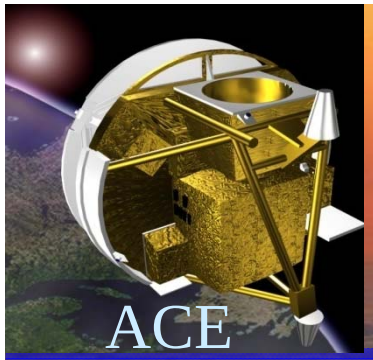

Version 4.0 CO_2

- CO_2 VMR in the pressure/temperature retrieval is fixed at low altitudes. The assumed rate of increase used in previous versions (1.50155 ppm/year) is too low. This carries through to VMR retrievals, affecting trends.
- From Geoff Toon: a subroutine that calculates CO_2 VMR as a function of latitude and time of year.
- Uses age of air to generate the vertical CO_2 profile.
- Much more sophisticated than what we have been using (i.e., a single profile shape for all locations and seasons)
- Assumes CO_2 VMR is increasing at a rate of ½ percent per year, which would put the current rate of increase around 1.9-1.95 ppm.

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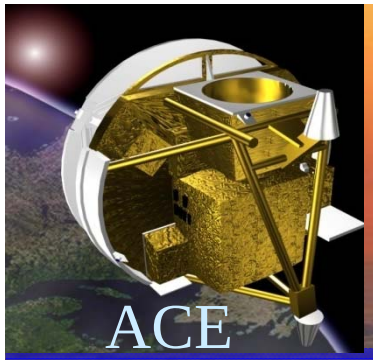
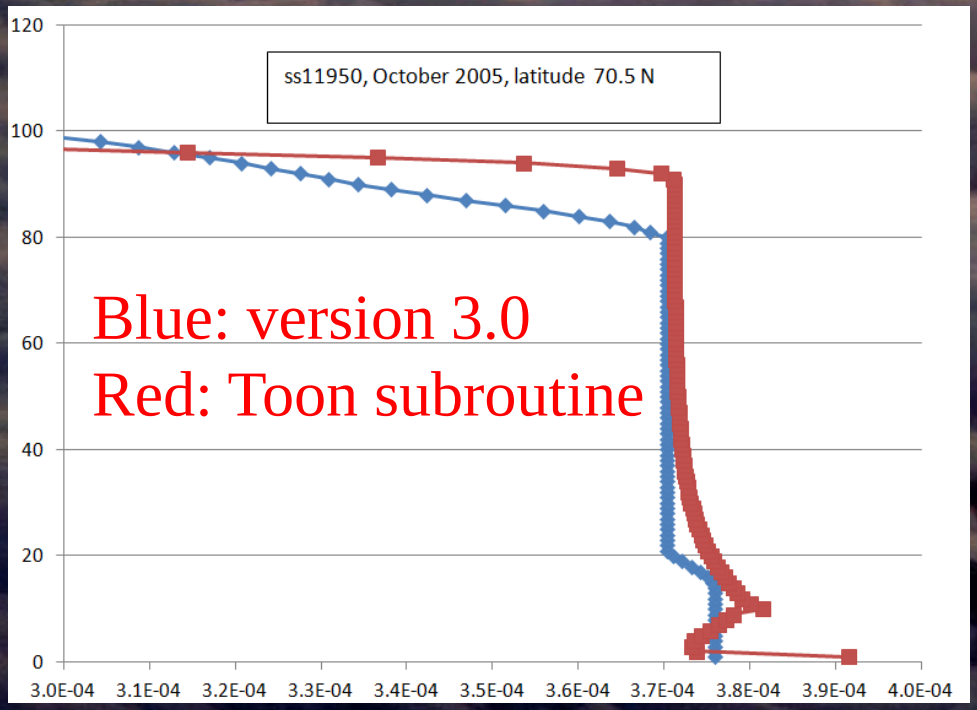
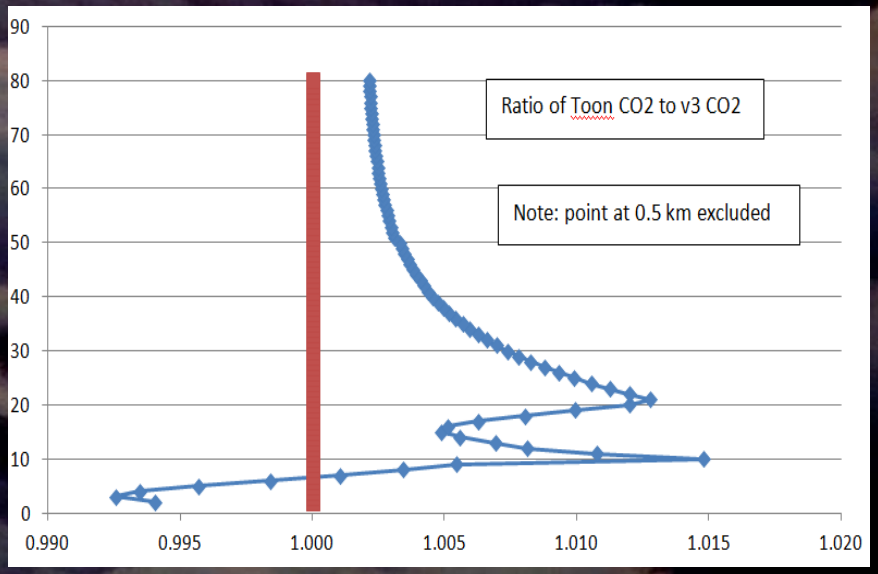


Polar Occultation, 2005

Differences of 0.5 to 1.5%. This will translate into similar differences in all of the retrieved VMRs.





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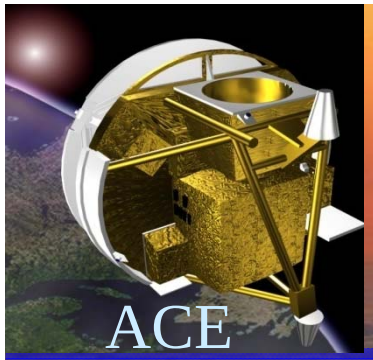
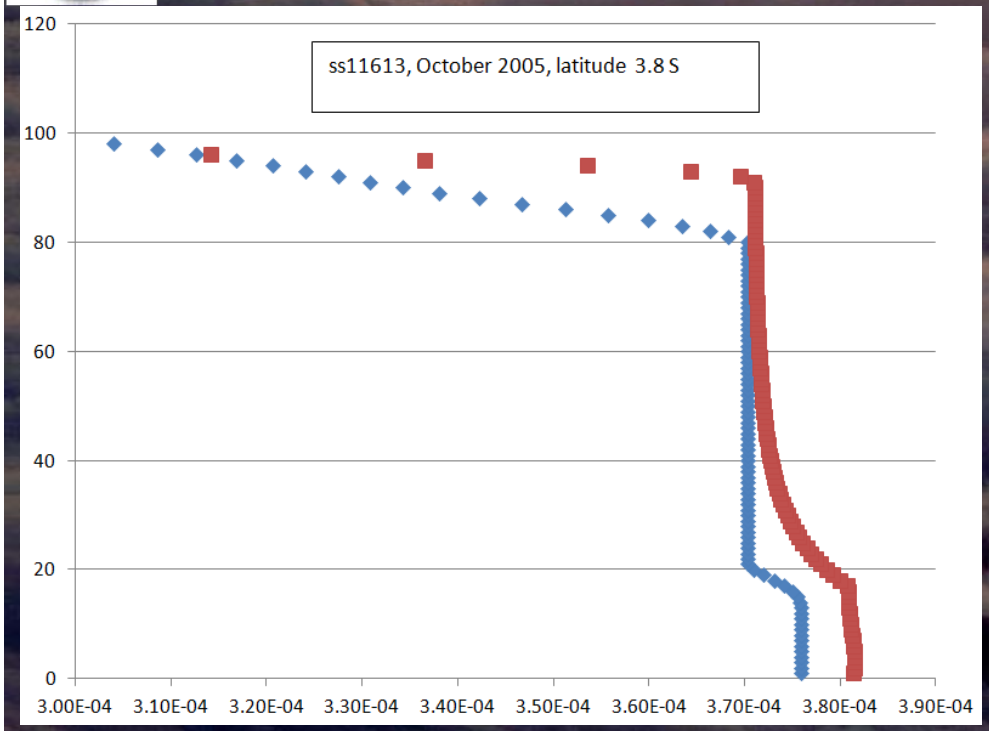
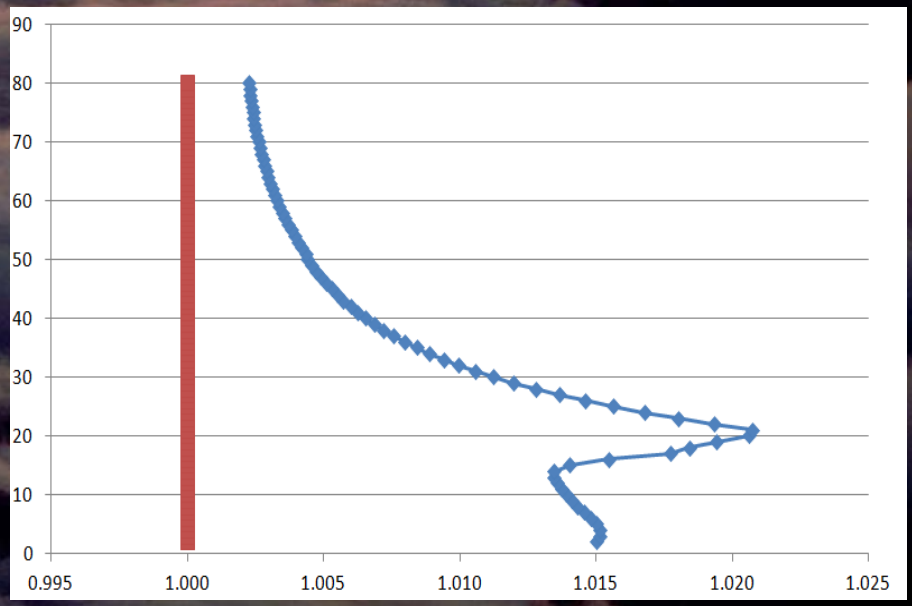


Tropical Occultation, 2005

Closer match in shape, but still significant differences.





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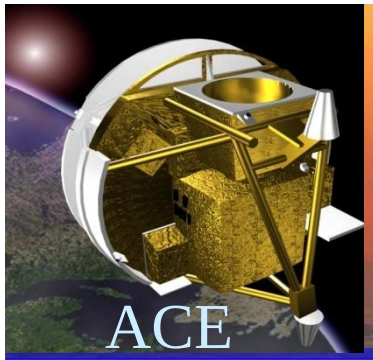
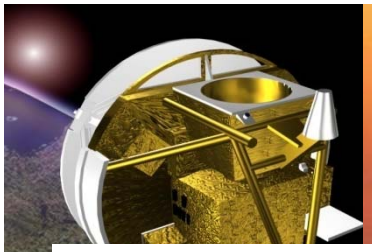
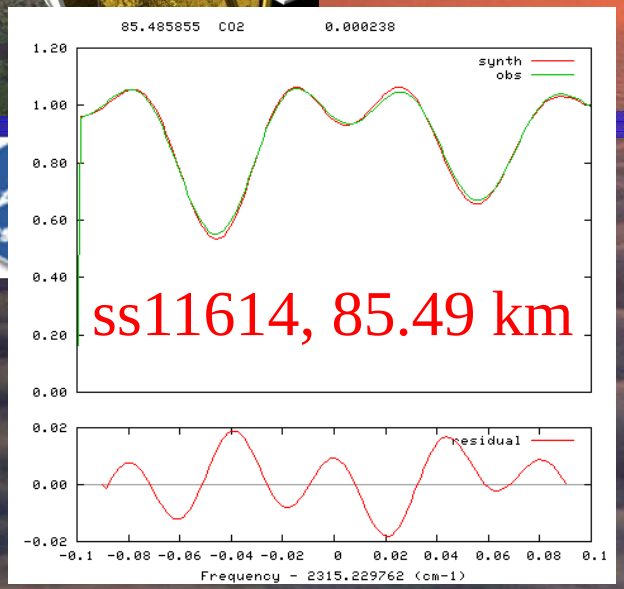


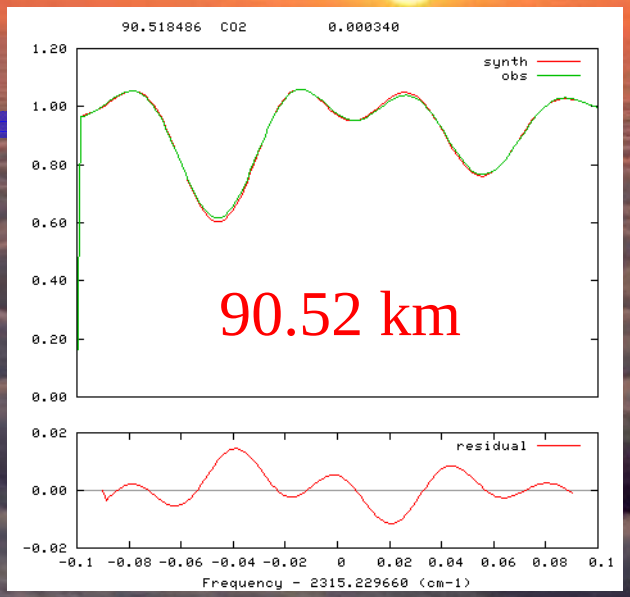
V4.0 Pressure/Temperature

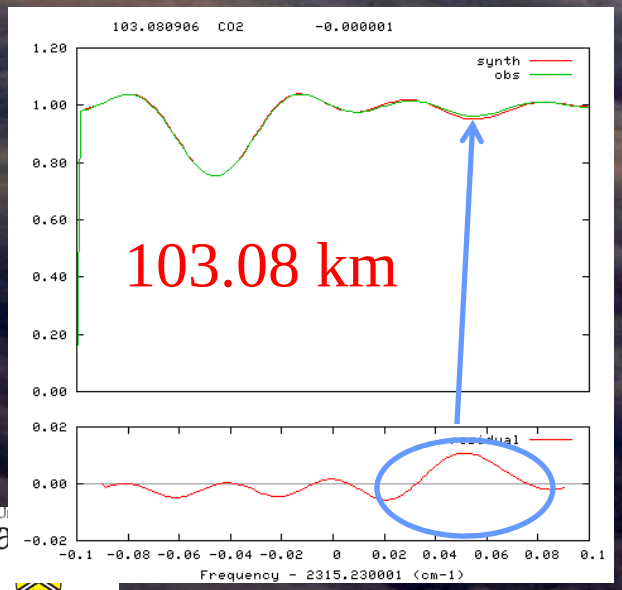
- Allowing interferers in the microwindows, allows the use of more high quality CO_2 lines and allows full P/T retrieval down to 5 km (15 km for v3.0).
- Interferers below ~60 km: $^{13}CO_2$, $^{18}O^{12}C^{16}O$, $^{17}O^{12}C^{16}O$, H_2O, O_3, N_2O, NO, OCS, and COF_2.
- Interferers above ~60 km: $^{13}CO_2$, $^{18}O^{12}C^{16}O$, $^{17}O^{12}C^{16}O$, and $^{18}O^{13}C^{16}O$
- Also treating a $^{16}O^{12}C^{16}O$ band with an excited lower vibrational state as an interferer. Retrieve a separate VMR profile for it above 80 km: infer vibrational temperature for the excited state.

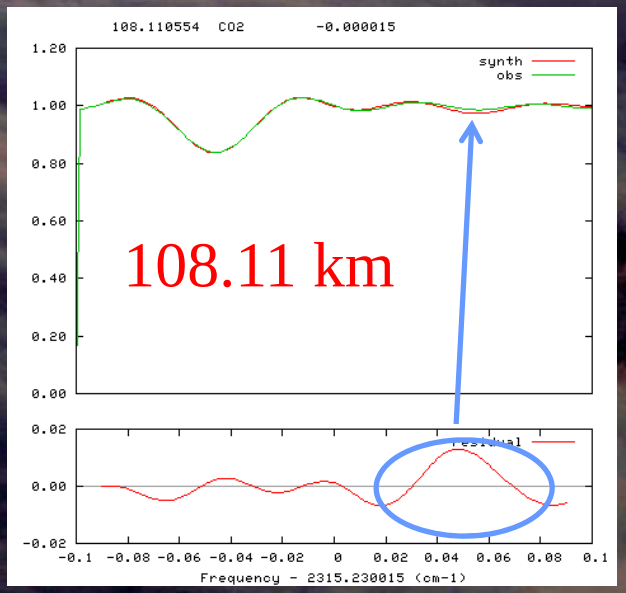


Non-local Thermodynamic Equilibrium Effects



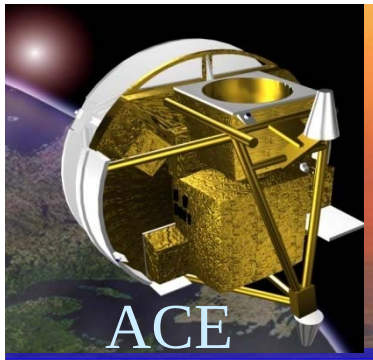






At lower altitudes, lines in $^{16}O^{12}C^{16}O$ with different lower state vibrations have comparable residuals.

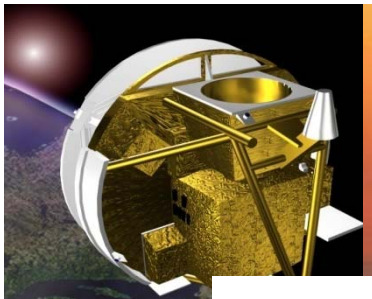
At higher altitudes, residuals for lines associated with excited vibrational states are quite large (percentagewise).



V4.0 P/T (continued)

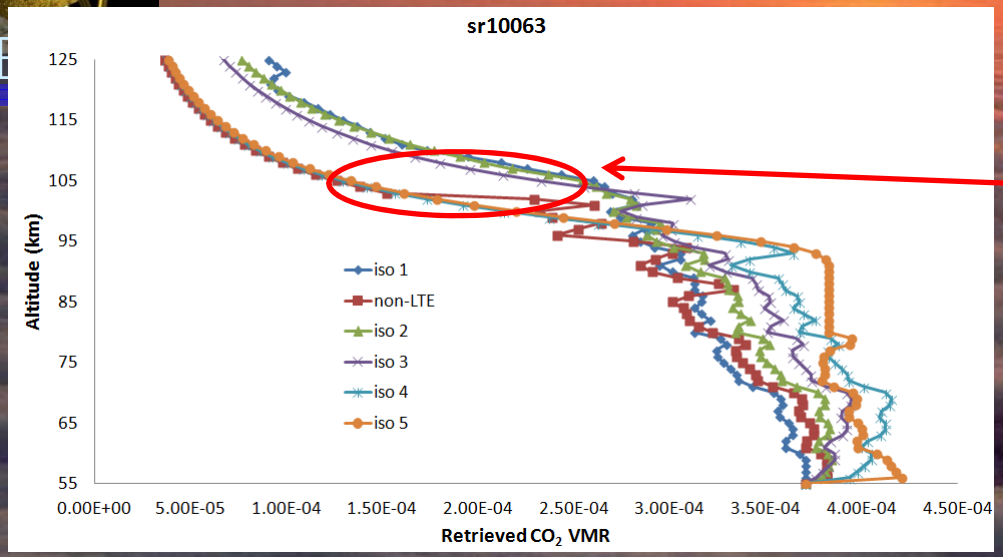
- CO_2 below ~60 km is fixed, while for altitudes above that CO_2 VMR is retrieved.
- Employed empirical function for high altitude CO_2 VMR in previous processing versions to suppress spikes.
- No empirical function for CO_2 VMR in version 4. Additional constraint required at high altitude. No regularization or explicit smoothing is performed.
- For CO_2 isotopologues 2, 3, 4, and 5, express the VMR profile as a linear function of altitude times that of the main (target) isotopologue:

$$VMR\_interferer(z) = (a + b*z) * VMR\_CO_2(z)$$

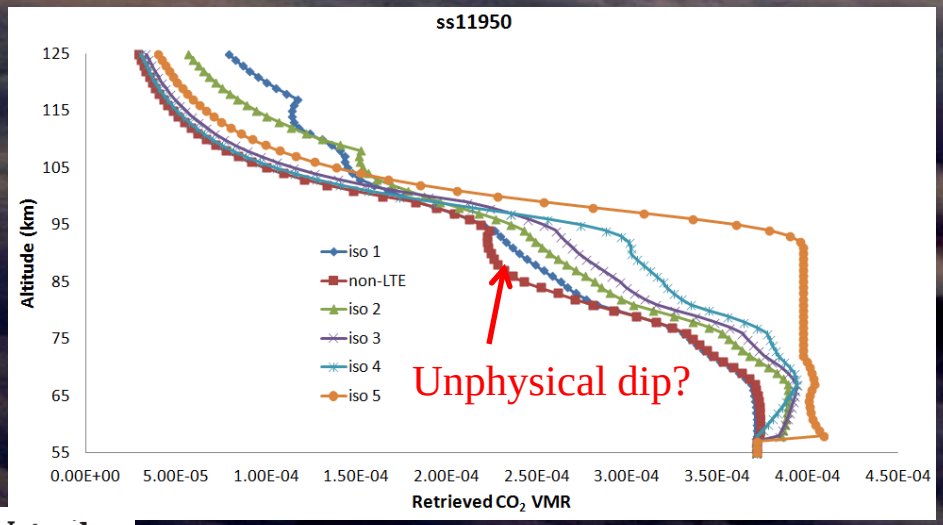


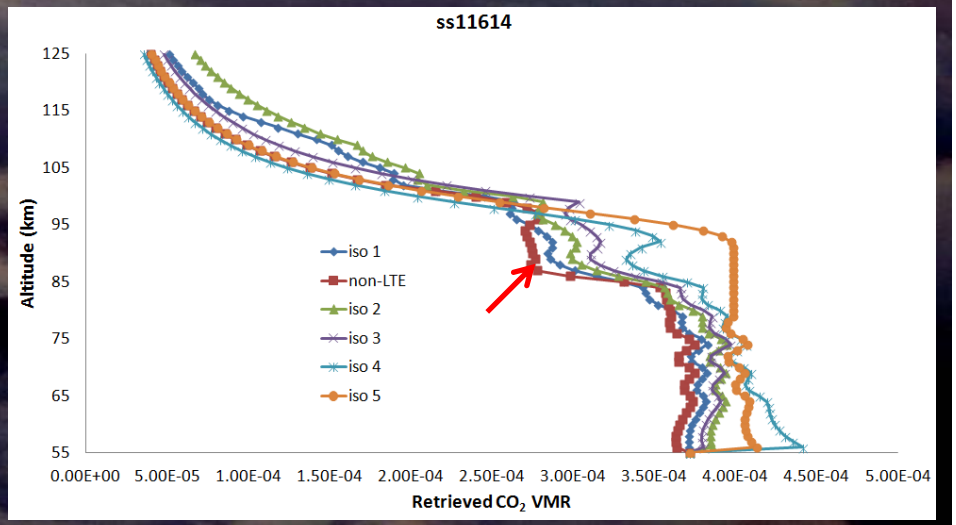
ACE

CO_2 VMR from P/T Retrieval



Strong non-LTE effects above ~100 km.





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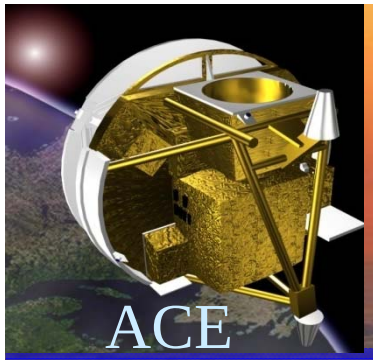

ACE-FTS Retrieved Molecules

- In ACE-FTS version 3.0 (37 molecules): CO_2, H_2O, O_3, N_2O, CO, CH_4, NO, NO_2, HNO_3, HF, HCl, $ClONO_2$, N_2O_5, CFC-11, CFC-12, OCS, HCN, CH_3Cl, CF_4, CCl_4, COF_2, C_2H_2, C_2H_6, CH_3OH, SF_6, HCOOH, HCFC-22, N_2, O_2, CFC-113, HCFC-141b, HCFC-142b, HNO_4, H_2O_2, H_2CO, $COCl_2$, and COClF
- New in version 4.0: HFC-23, PAN, acetone, CH_3CN, HFC-134a. SO_2 will also be retrieved in v4.0, with good results in enhanced conditions (volcanic plumes), but current SO_2 levels are too low for good results under background conditions

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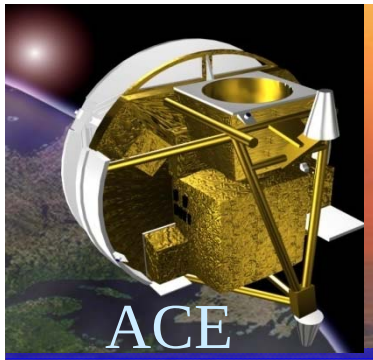
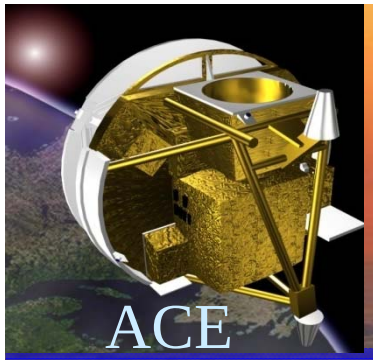


Isotopologues

- In ACE-FTS version 3.0 (21 subsidiary isotopologues): $H_2{}^{18}O$, $H_2{}^{17}O$, HDO, $^{13}CO_2$, $^{18}O^{12}C^{16}O$, $^{17}O^{12}C^{16}O$, $^{18}O^{13}C^{16}O$, $^{18}O^{16}O^{16}O$, $^{16}O^{18}O^{16}O$, $^{16}O^{17}O^{16}O$, $^{14}N^{15}N^{16}O$, $^{15}N^{14}N^{16}O$, $N_2{}^{18}O$, $N_2{}^{17}O$, $^{13}C^{16}O$, $^{12}C^{18}O$, $^{12}C^{17}O$, $^{13}CH_4$, CH_3D, $OC^{34}S$, and $O^{13}CS$.
- New in version 4.0: $H^{15}NO_3$.



Minimizing H_2O Impact

- A number of molecules have broad spectral features in the ACE-FTS (CFC-11, N_2O_5,...).
- H_2O lines appear as interferers in many of these broad spectral features.
- In previous processing versions, bad residuals from H_2O lines may have degraded the retrieval quality.
- In v4.0, use sets of interconnected microwindows across the broad spectral feature (rather than a single large window) to avoid strong H_2O lines.

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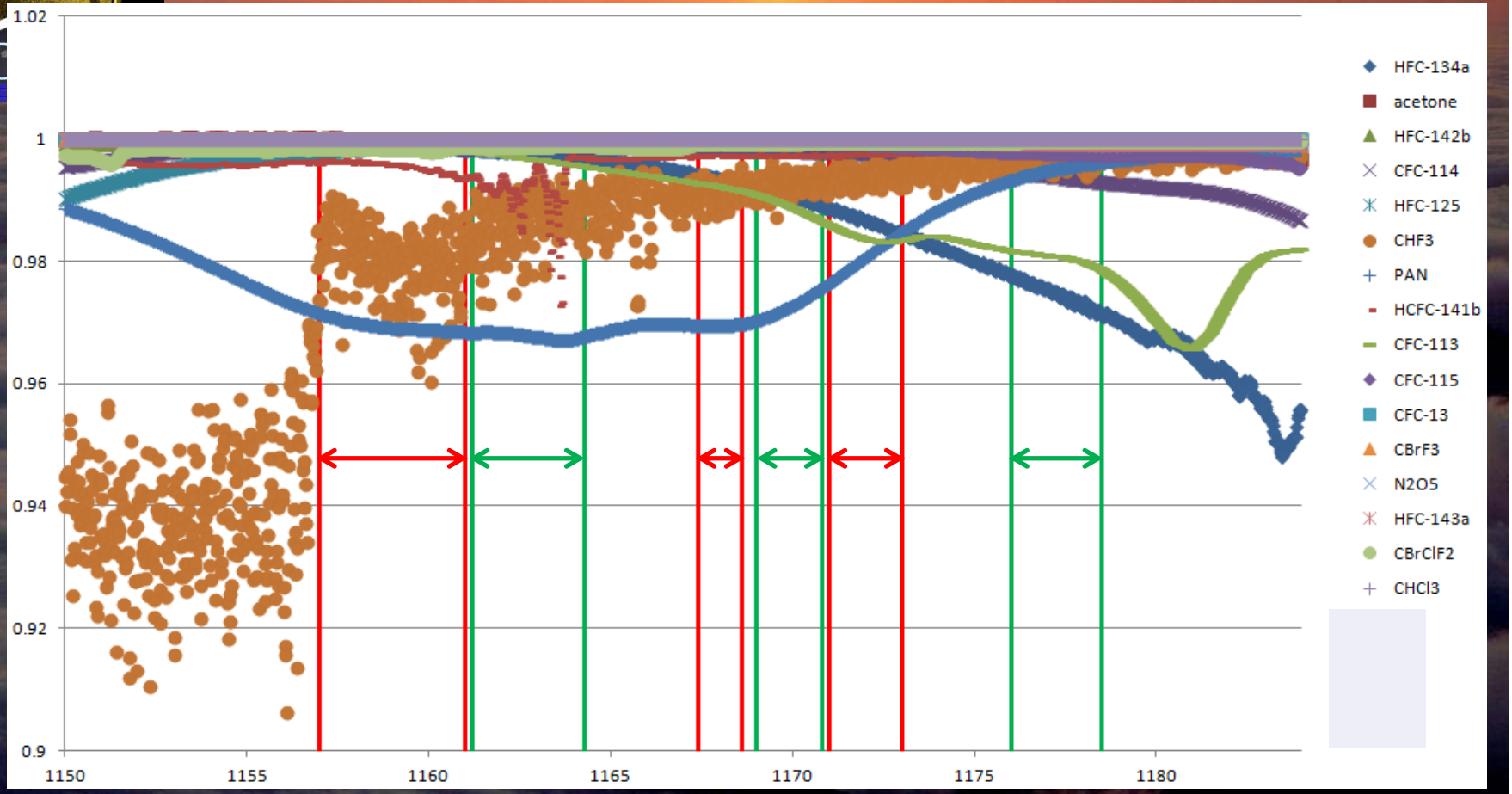
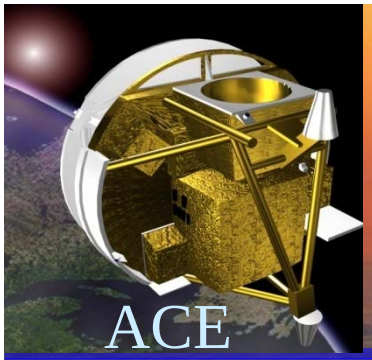
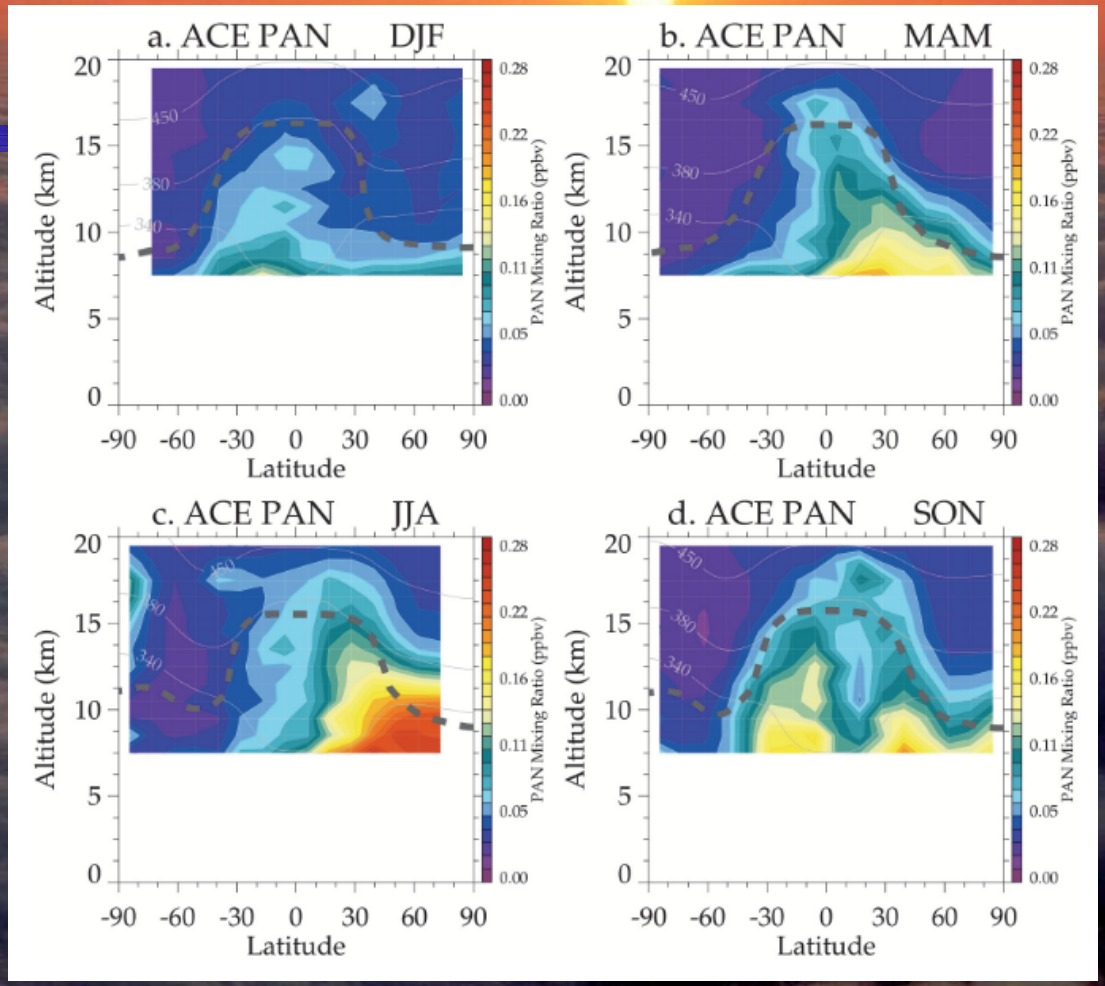
Peroxyacetyl Nitrate (PAN)



6 microwindows, selected to avoid bad residuals from H_2O lines (or other sources), with a common set of baseline fitting parameters (scale and slope) for all of the microwindows.



PAN First Results



K. A. Tereszchuk *et al.*, Observations of peroxyacetyl nitrate (PAN) in the upper troposphere by the Atmospheric Chemistry Experiment-Fourier Transform Spectrometer (ACE-FTS), Atmos. Chem. Phys., **13**, 5601-5613 (2013).

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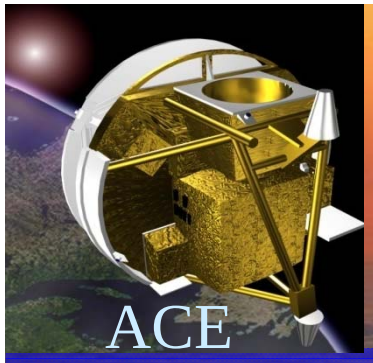
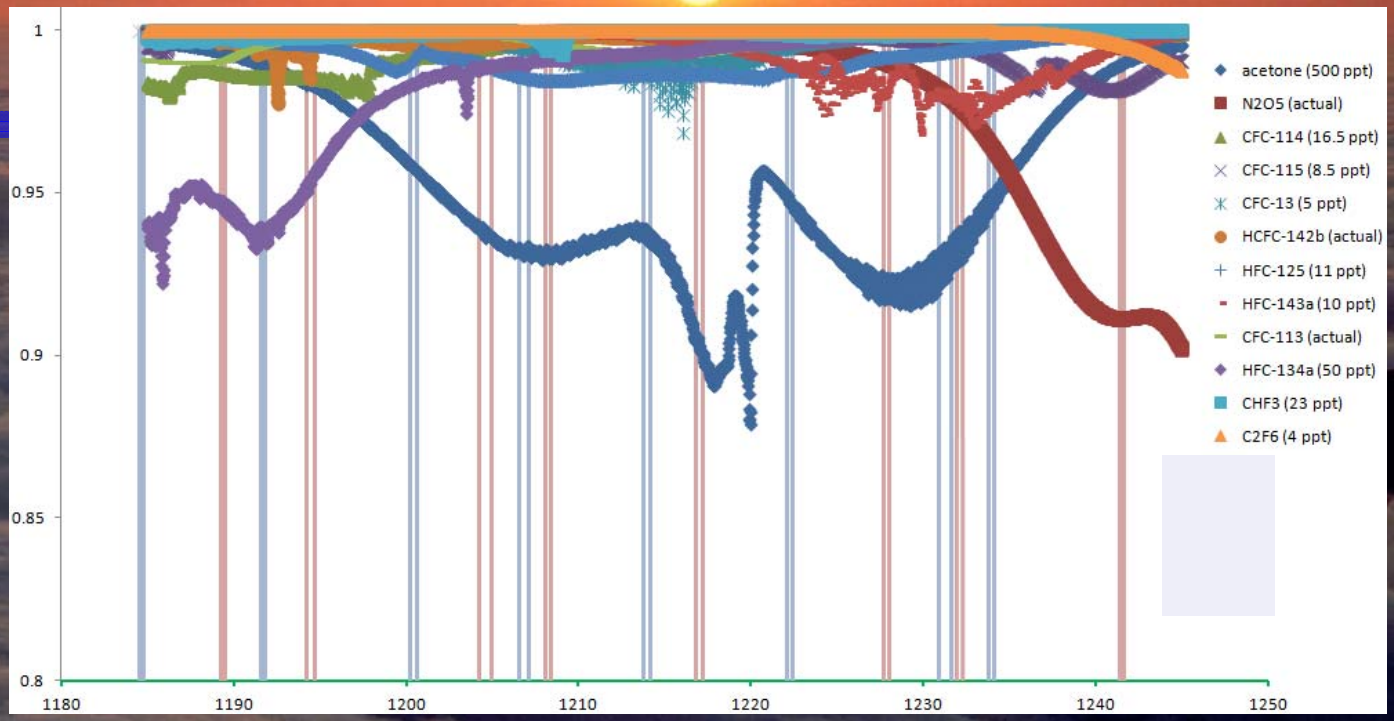
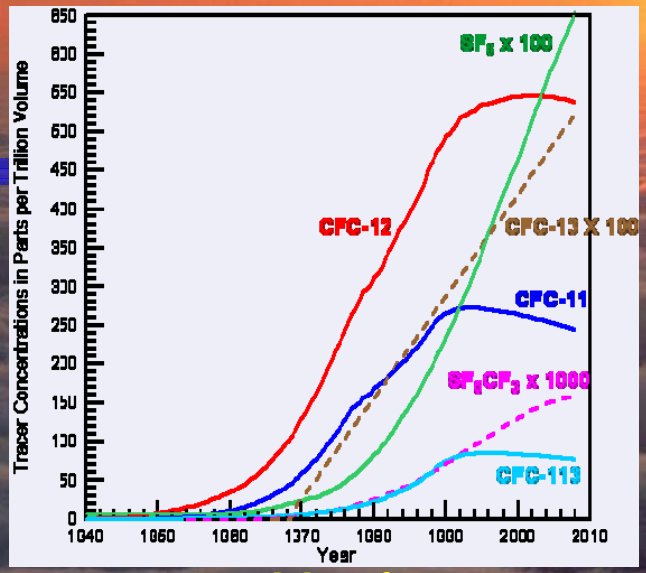


Acetone

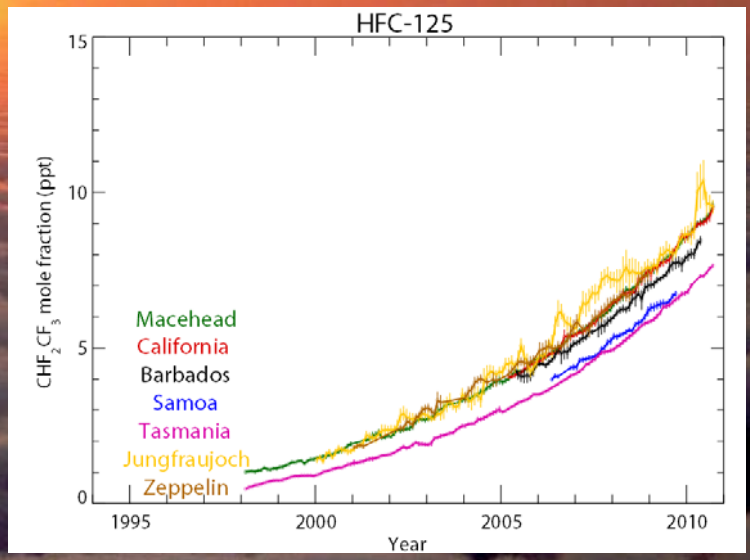


- In the acetone retrieval, insufficient information to properly determine a number of interferers, so they are fixed in the retrieval: CFC-12, HCFC-22, CFC-13, CFC-114, CFC-115, HFC-125, HFC-143a, $CBrF_3$.
- Need to account for trends of the interferers being fixed.

Trends in Interferers

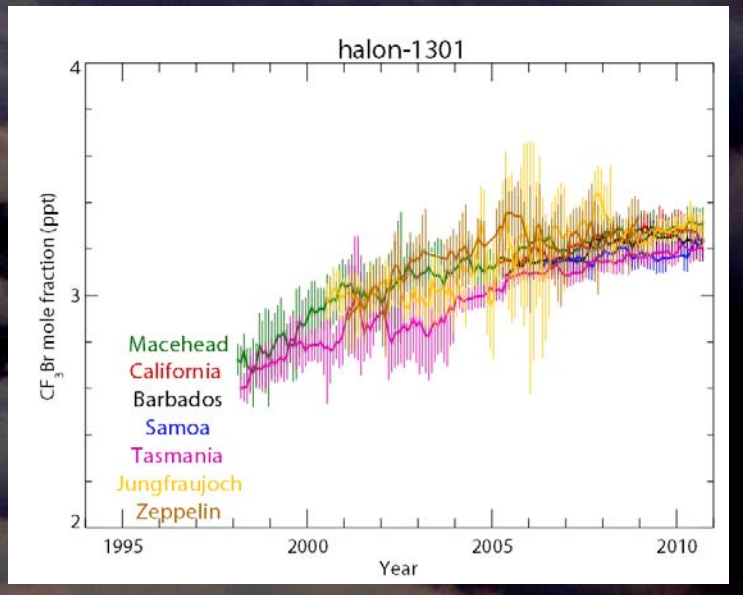


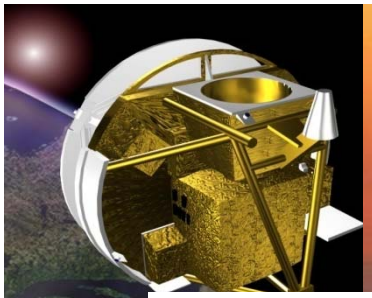
USGS: http://water.usgs.gov/lab/software/air_curve/



AGAGE: http://agage.eas.gatech.edu/data.htm

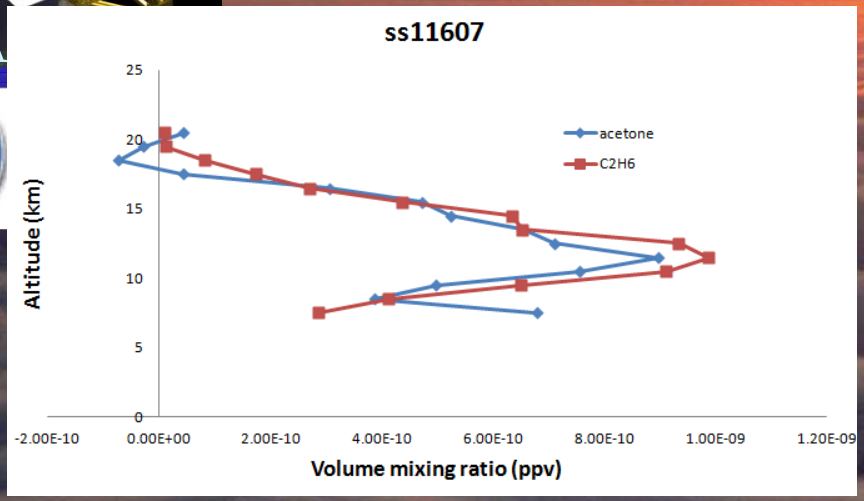
- Trends for CFC-114, CFC-115, HFC-143a, etc., taken from information in the WMO Scientific Assessment of Ozone Depletion: http://www.esrl.noaa.gov/csd/assessments/ozone/2010/report.html
- Often hemispheric differences.

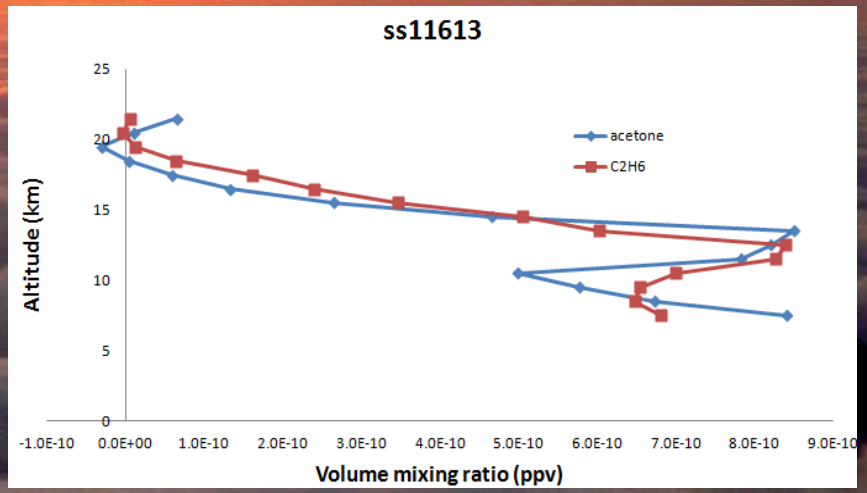




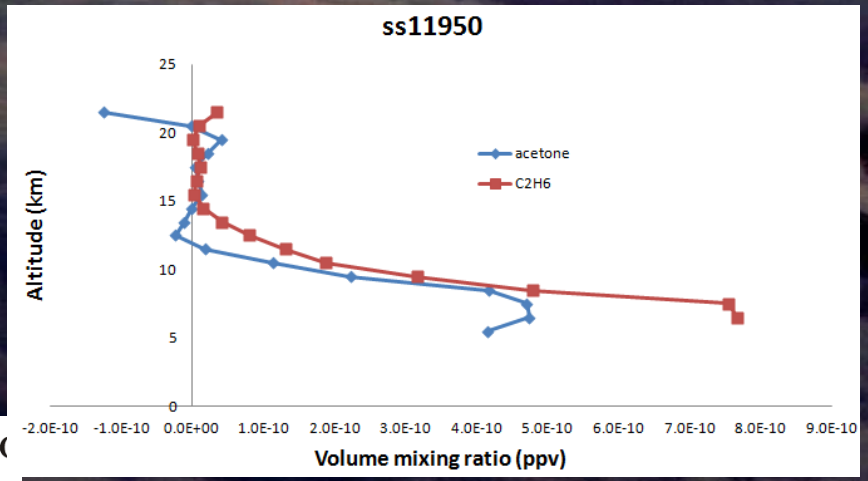
Acetone Retrievals

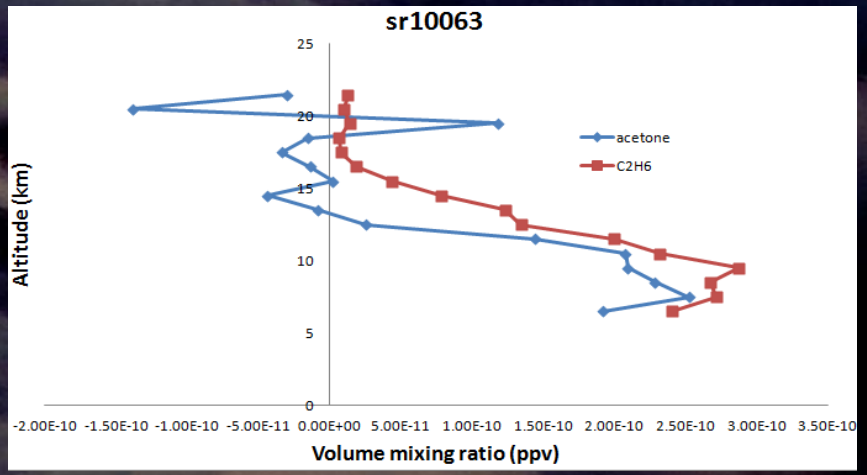
A



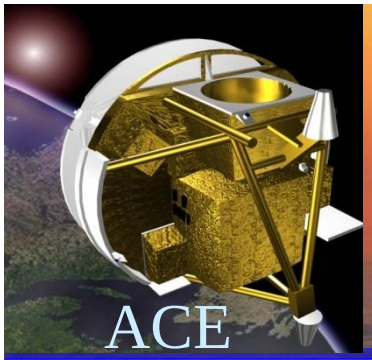


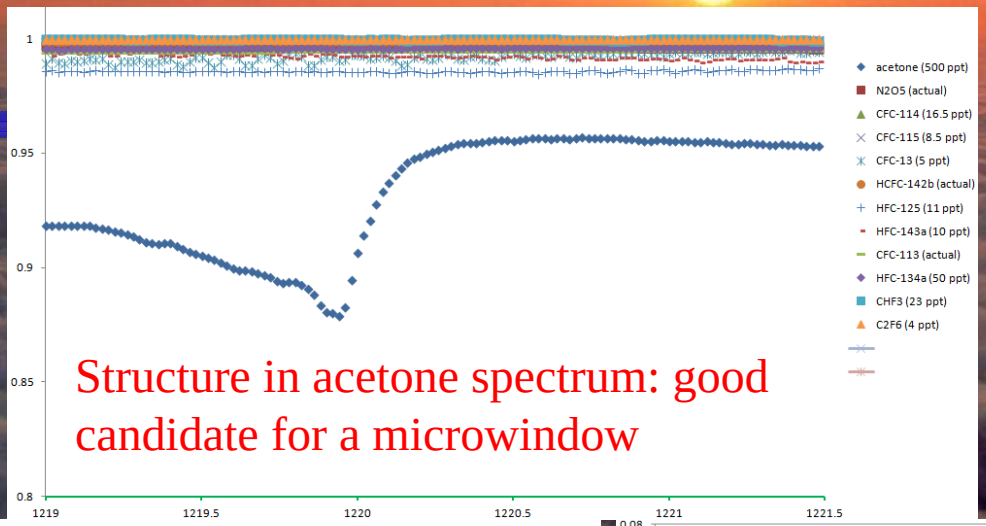
Strong correlation with C_2H_6 (both are biomass burning products)



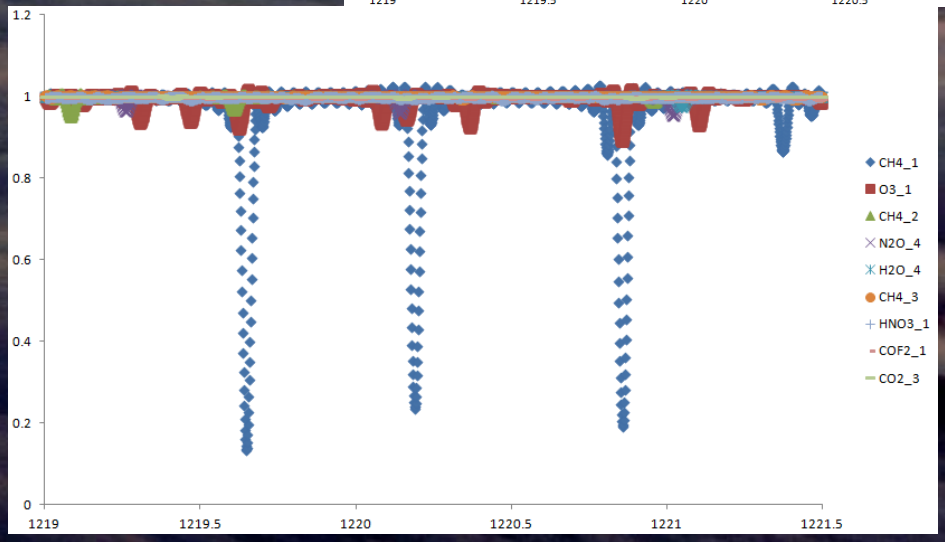


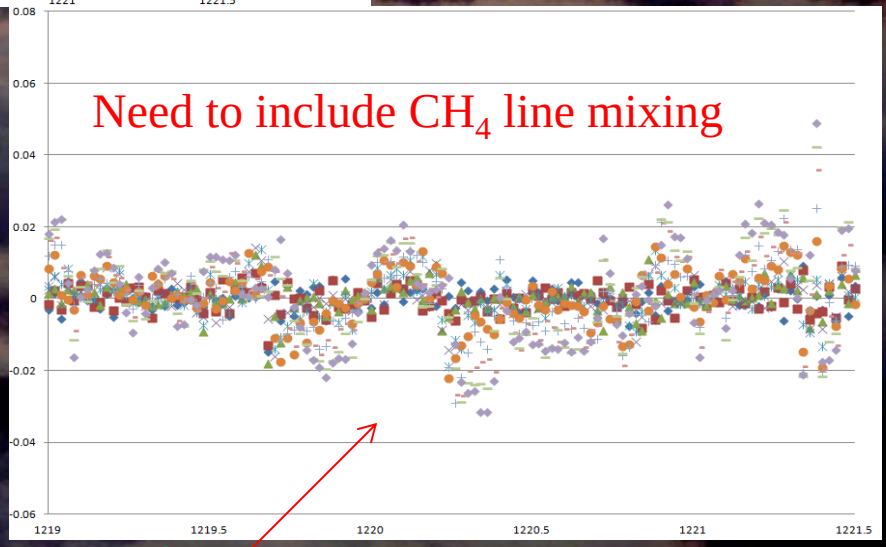
CH_4 Line Mixing





Structure in acetone spectrum: good candidate for a microwindow





Need to include CH_4 line mixing

Residuals for all analyzed measurements plotted simultaneously on one graph

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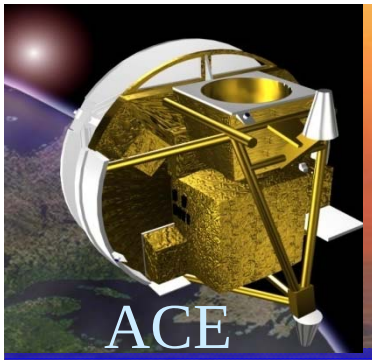
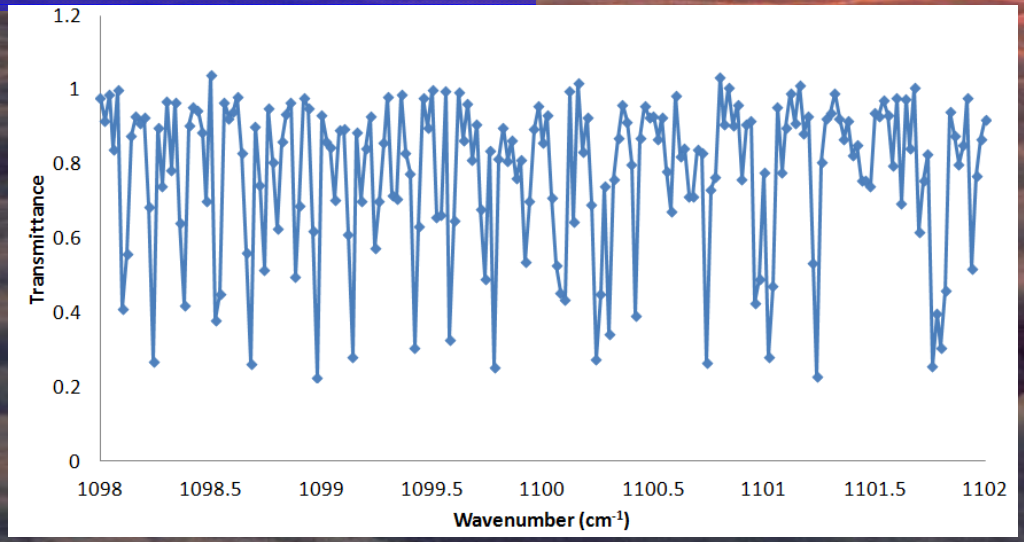
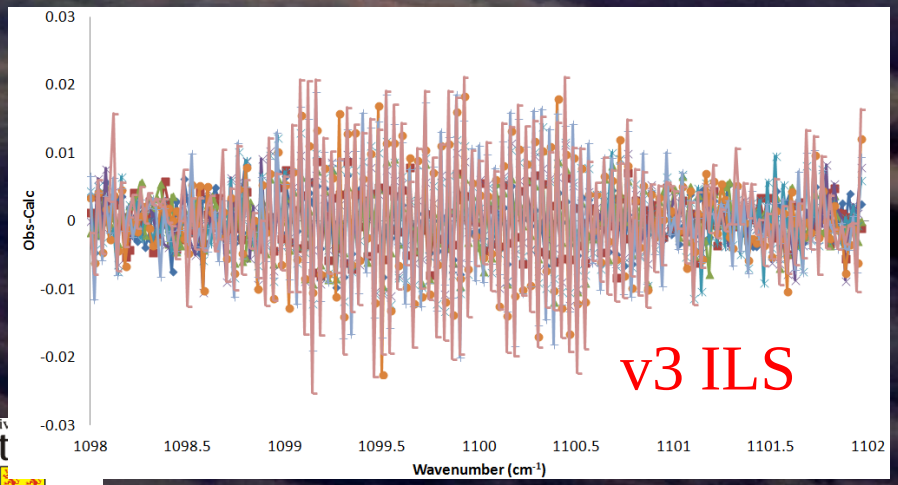
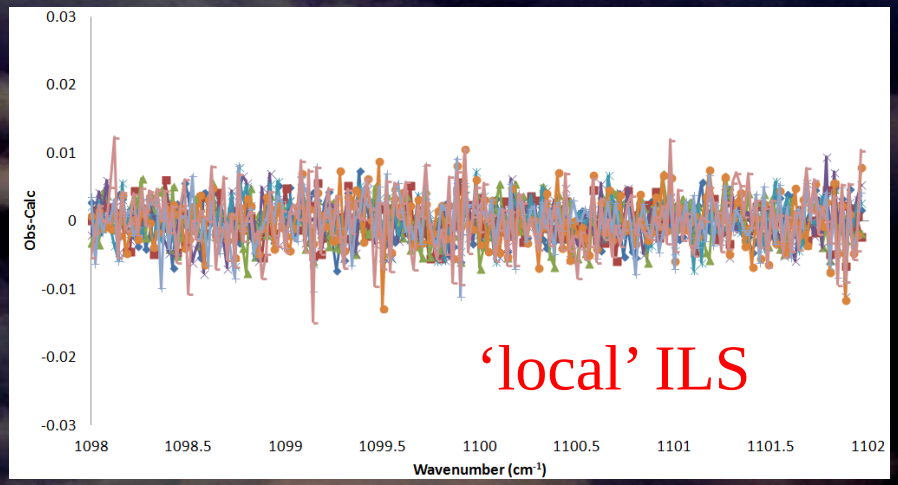


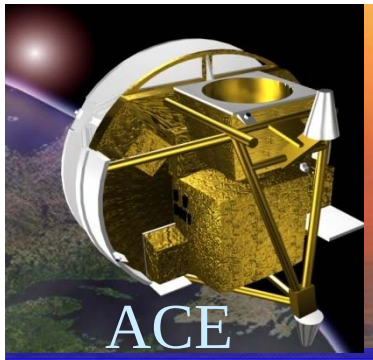
Instrumental Line Shape

Previously used only clean lines (isolated with minimal interferences) to determine ACE-FTS ILS. For v4, use "messy" regions with a strong sensitivity to the ILS to refine the model. Proper treatment of wavenumber dependence is key.









Summary

- Versions 2.5 and 3.5 are in progress, replacing results from October 2010 onward.
- Looking to start version 4 sometime this year.
  - New CO_2 VMR a priori.
  - New spectroscopic data (HITRAN 2012).
  - Improved model of the ACE-FTS instrumental line shape
  - Including weak interferers previously ignored (CFC-114, etc.).
  - Minimizing pollution of retrievals for molecules with broad absorption features from H_2O, neglected CH_4 line mixing, etc.
  - New molecules (CHF_3, PAN, acetone, CH_3CN, HFC-134a, $H^{15}NO_3$)
- Reviewing microwindow selections for all targets. High altitude P/T ready. Looking to clean up residual solar features for low altitude portion of P/T retrievals.

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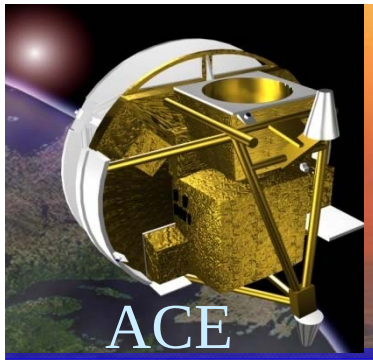

Acknowledgment

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