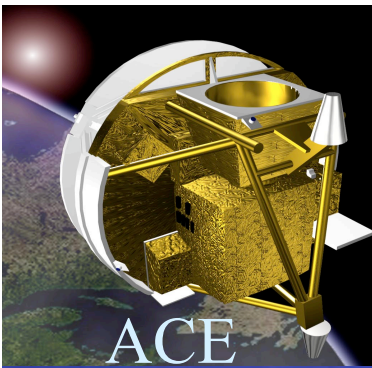


The Atmospheric Chemistry Experiment (ACE): Status and Latest Results

Chris Boone and Peter Bernath

5th Limb Conference and Workshop
November 16, 2009



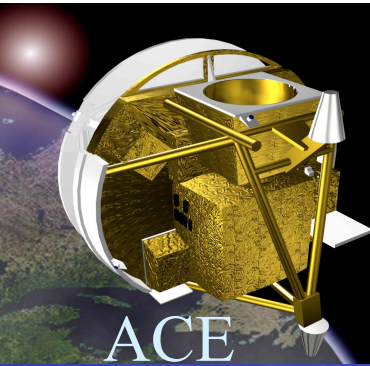


ACE Soldiering On

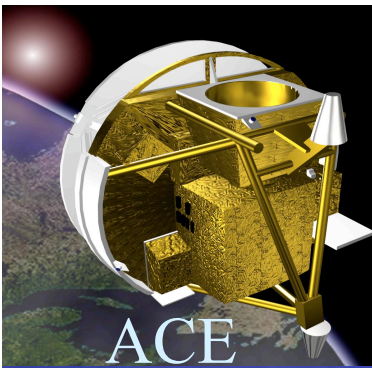


- ACE launched August 2003
- Science operations began February 2004
- Satellite and all instruments continue nominal operation.
- Version 3.0 of the data processing began in October 2009.
- Version 2.2 planned for public release.

Instruments



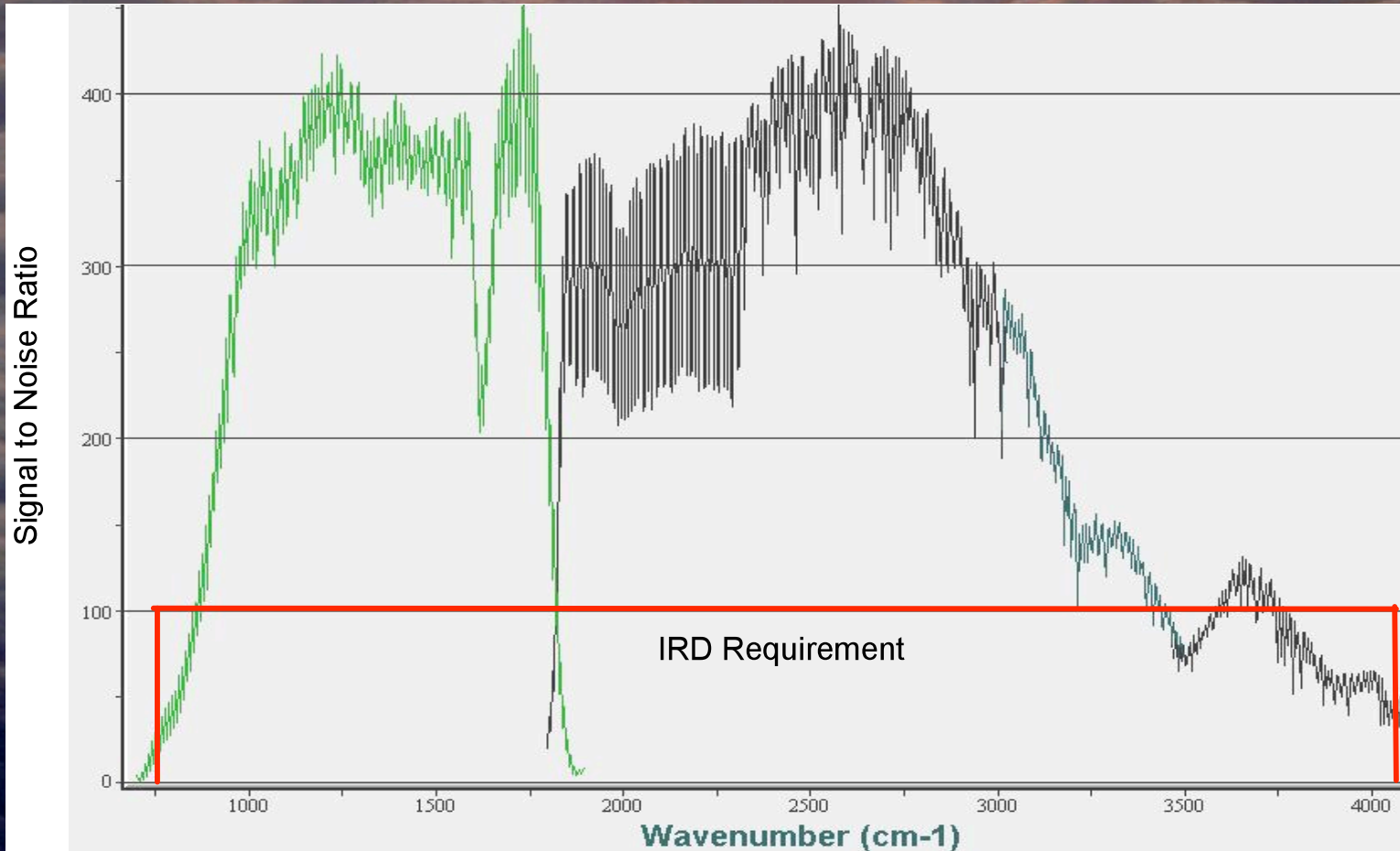
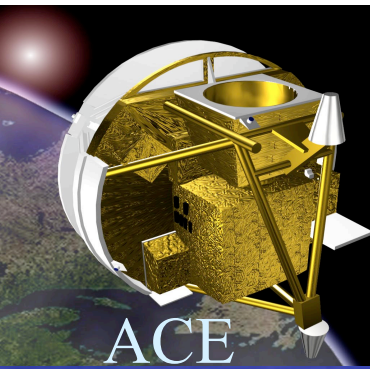
- Infrared Fourier Transform Spectrometer operating between 2 and 13 microns with a resolution of 0.02 cm^{-1} ($\pm 25 \text{ cm MPD}$)
- 2-channel visible/near infrared Imagers, operating at 0.525 and 1.02 microns
- UV / Visible spectrometer (MAESTRO) 0.285 to 1.03 microns, resolution $\sim 1\text{-}2 \text{ nm}$
- Suntracker
- Startracker

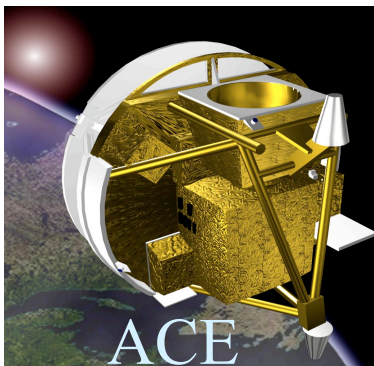


ACE-FTS (ABB-Bomem)



SNR

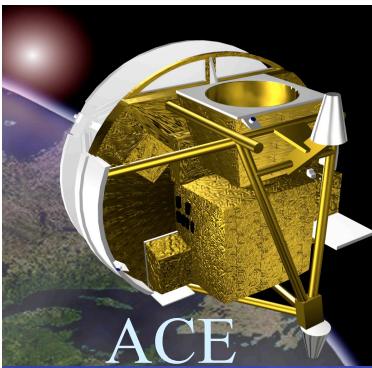




Molecules



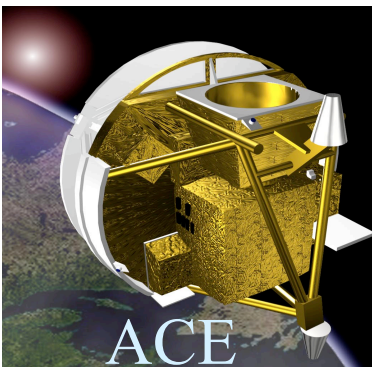
- In ACE-FTS version 3.0 (37 molecules):
CO₂, H₂O, O₃, N₂O, CO, CH₄, NO, NO₂,
HNO₃, HF, HCl, ClONO₂, N₂O₅, CFC-11,
CFC-12, OCS, HCN, CH₃Cl, CF₄, CCl₄, COF₂,
C₂H₂, C₂H₆, CH₃OH, SF₆, HCOOH, HCFC-22,
N₂, O₂, CFC-113, HCFC-141b, HCFC-142b,
HNO₄, H₂O₂, H₂CO, COCl₂, COClF
- Future products?: HFC-134a, HFC-23,
C₂H₄, SO₂, NH₃, PAN, acetone, propane,
BrONO₂, ...



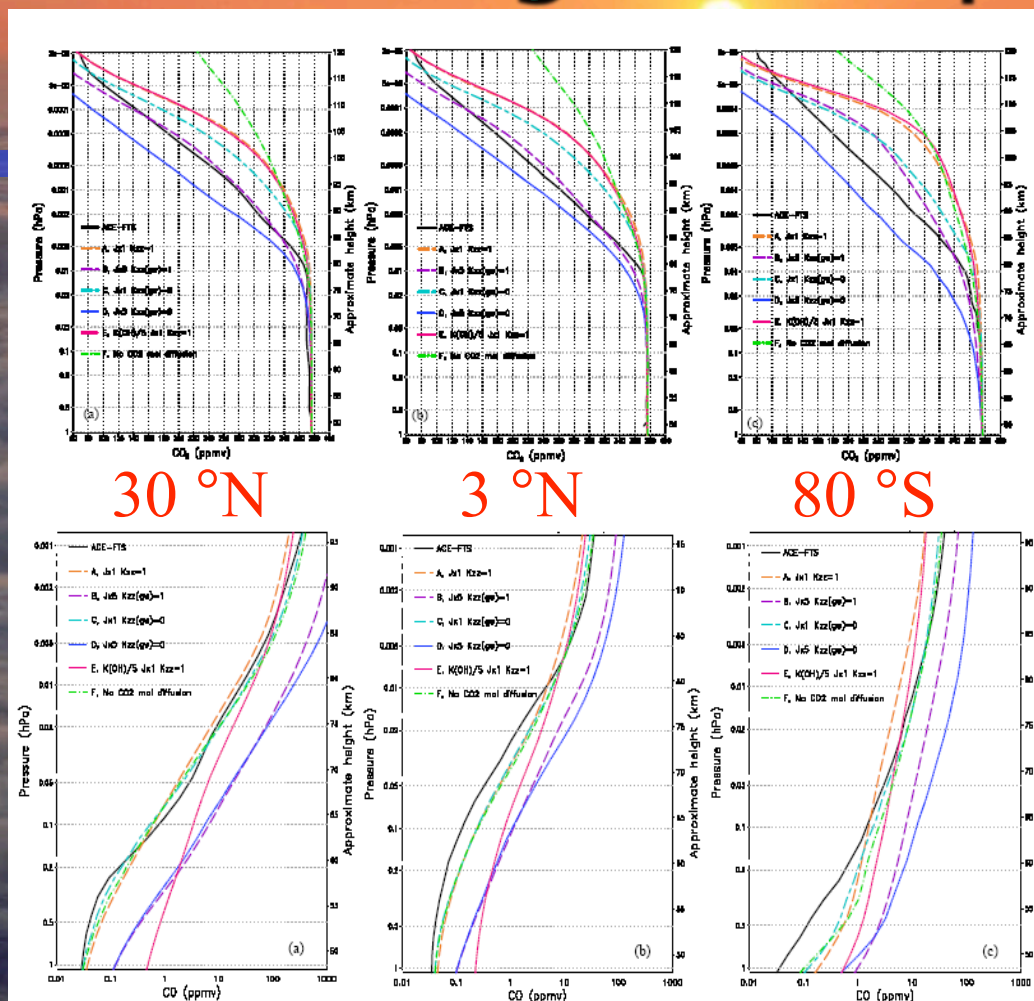
Isotopologues



- In ACE-FTS version 3.0 (23 subsidiary isotopologues): H_2^{18}O , H_2^{17}O , HDO , $^{13}\text{CO}_2$, $^{18}\text{O}^{12}\text{C}^{16}\text{O}$, $^{17}\text{O}^{12}\text{C}^{16}\text{O}$, $^{18}\text{O}^{13}\text{C}^{16}\text{O}$, $^{18}\text{O}^{16}\text{O}^{16}\text{O}$, $^{16}\text{O}^{18}\text{O}^{16}\text{O}$, $^{17}\text{O}^{16}\text{O}^{16}\text{O}$, $^{16}\text{O}^{17}\text{O}^{16}\text{O}$, $^{14}\text{N}^{15}\text{N}^{16}\text{O}$, $^{15}\text{N}^{14}\text{N}^{16}\text{O}$, N_2^{18}O , N_2^{17}O , $^{13}\text{C}^{16}\text{O}$, $^{12}\text{C}^{18}\text{O}$, $^{12}\text{C}^{17}\text{O}$, $^{13}\text{CH}_4$, CH_3D , OC^{34}S , O^{13}CS , OC^{33}S



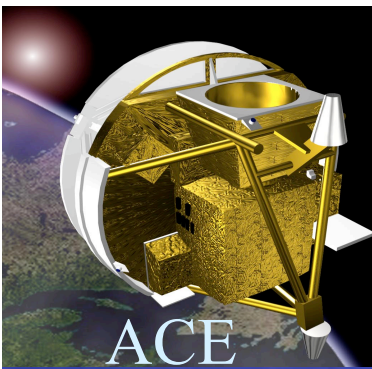
Modeling mesospheric CO₂



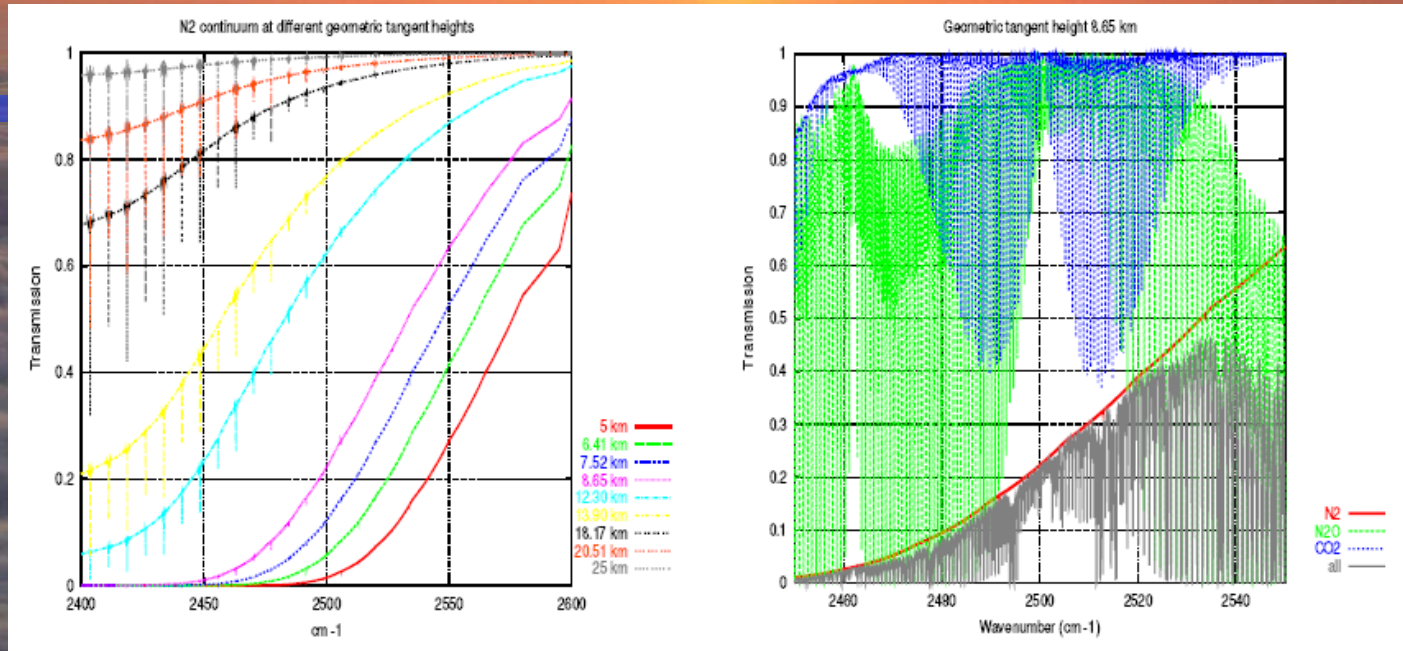
In the CMAM model, cannot reconcile CO₂ and CO.

Similar study underway using the WACCAM model.

First multi-year occultation observations of CO₂ in the MLT by ACE satellite: observations and analysis using the extended CMAM, S. R. Beagley, C. D. Boone, V. I. Fomichev, J. J. Jin, K. Semeniuk, J. C. McConnell, and P. F. Bernath, Atmos. Chem. Phys. Discussions, 9, 11551-11587 (2009).



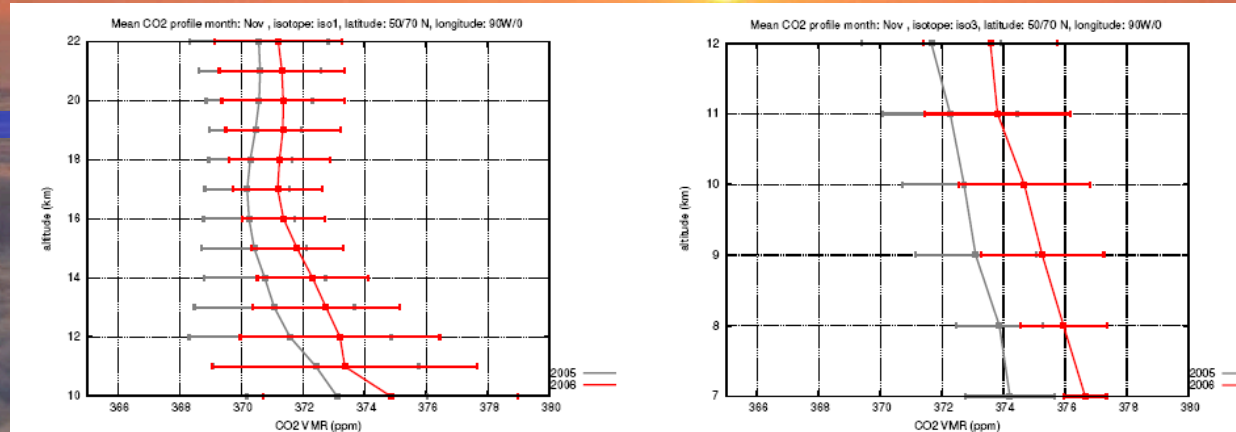
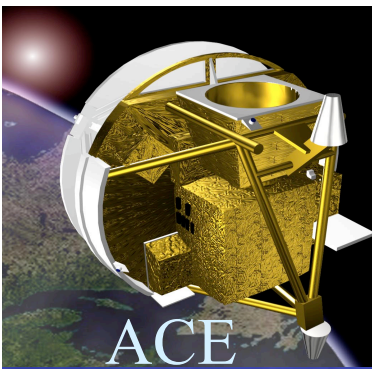
Retrieving UT/LS CO₂



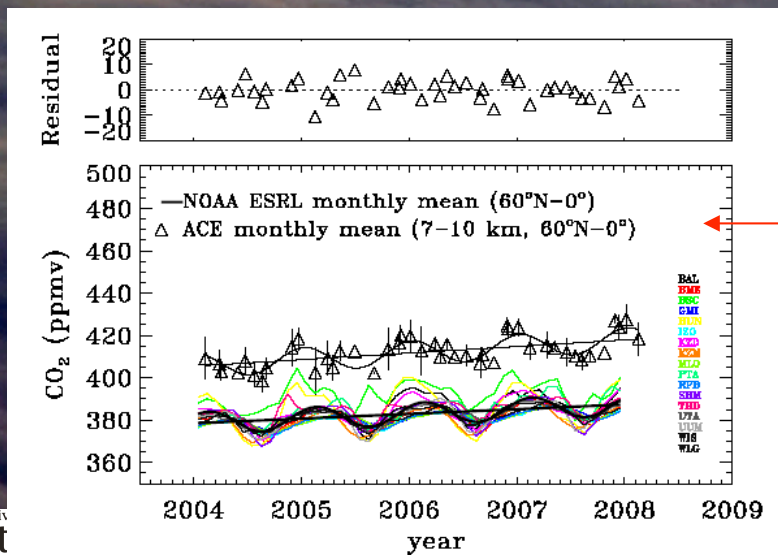
Technical Note: Feasibility of CO₂ profile retrieval from limb viewing solar occultation made by the ACE-FTS instrument, P. Y. Foucher, A. Chedin, G. Dufour, V. Capelle, C. D. Boone, and P. Bernath, Atmos. Chem. Phys., 9, 2873-2890 (2009).

- Analysis of CO₂ spectral features used to determine ACE-FTS pressure, temperature, and tangent heights, with CO₂ VMR fixed to an assumed value.
- Retrieving CO₂ would be a circular problem
- Generate independent tangent heights using the N₂ continuum.

Two independent studies



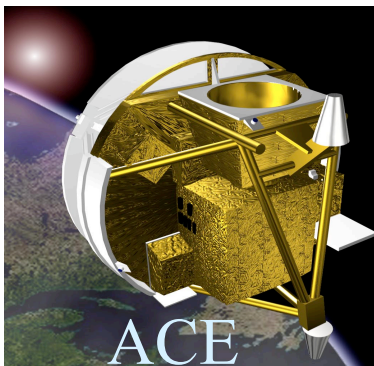
Technical Note: Feasibility of CO₂ profile retrieval from limb viewing solar occultation made by the ACE-FTS instrument, P. Y. Foucher, A. Chedin, G. Dufour, V. Capelle, C. D. Boone, and P. Bernath, Atmos. Chem. Phys., 9, 2873-2890 (2009).



One study: CO₂ isotopologue 1 above 10 km, OC¹⁸O below 12 km, scale latter results. Generate profiles.

Other study: only use OC¹⁸O lines and acknowledge expected offset. Generate average value for 7-10 km.

Carbon dioxide (CO₂) retrievals from Atmospheric Chemistry Experiment (ACE) solar occultation measurements, Curtis P. Rinsland, Linda Chiou, Chris Boone, and Peter Bernath, J. Geophys. Res., in press



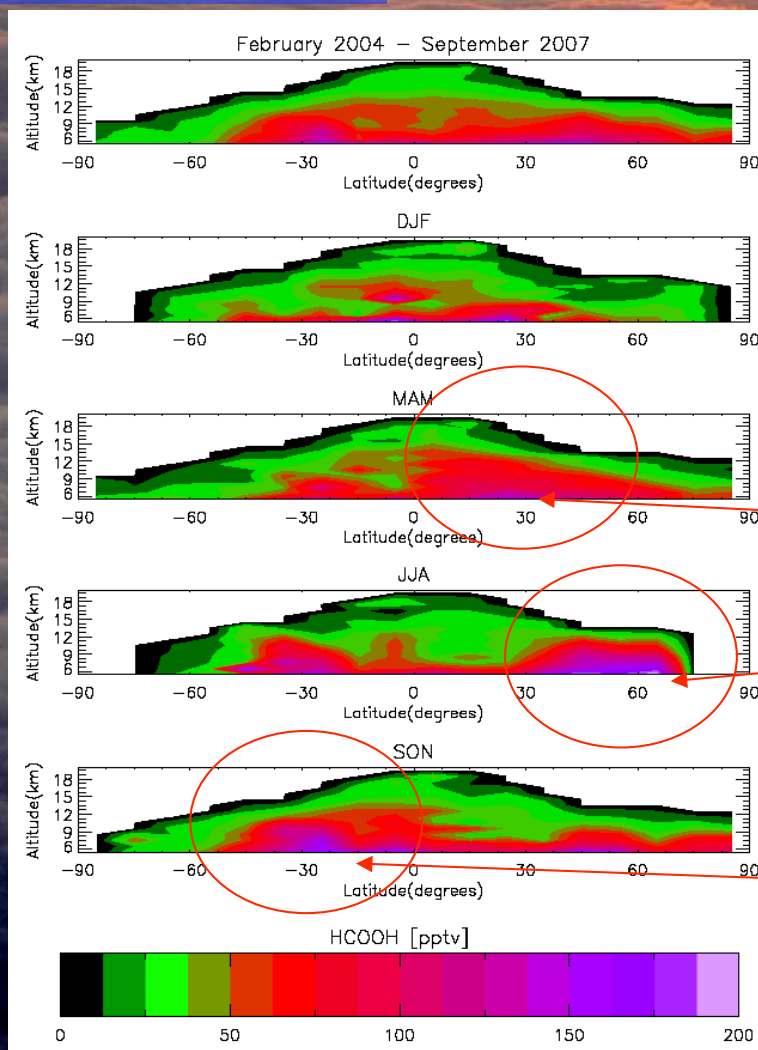
HCOOH

Global distribution of upper tropospheric formic acid from the ACE-FTS, G. Gonzalez Abad, P. F. Bernath, C. D. Boone, S. D. McLeod, G. L. Manney, and G. C. Toon, *Atmos. Chem. Phys.*, 9, 8039-8047 (2009)

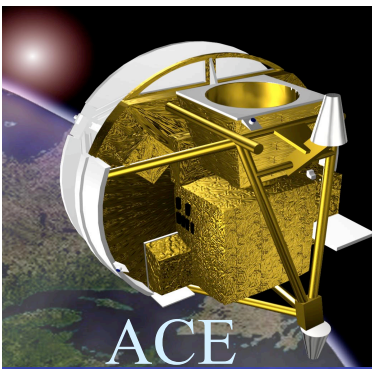
Growing season

Boreal fires + plant growth?
Challenge for models.

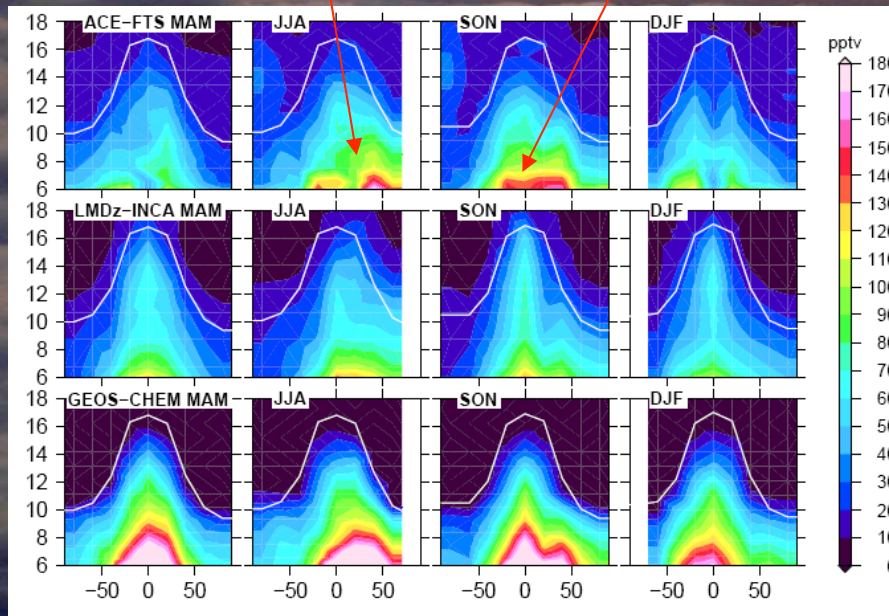
Biomass burning



H₂CO

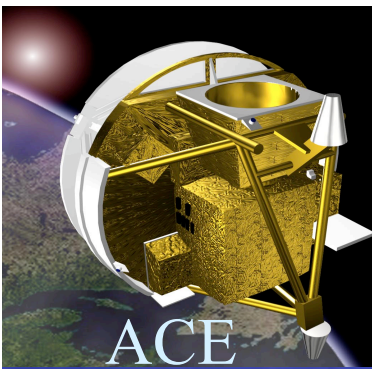


Growing season Biomass burning

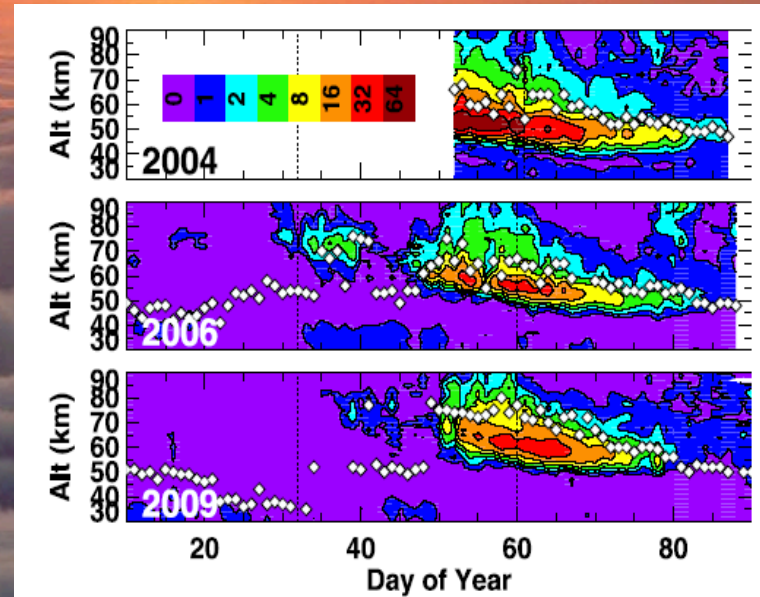
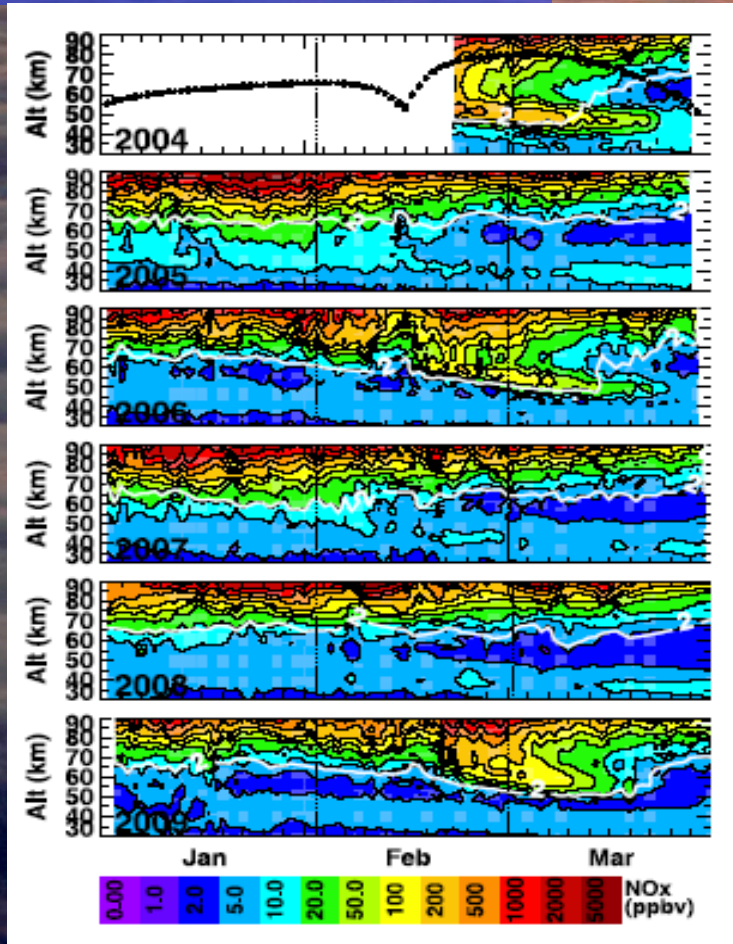


A good test for chemistry transport models. H₂CO in the upper troposphere is mostly from secondary production. Need to properly model precursors, transport, and chemistry. Current models are not yet up to the task.

Global upper-tropospheric formaldehyde: seasonal cycles observed by the ACE-FTS satellite instrument, G. Dufour, S. Szopa, M. P. Barkley, C. D. Boone, A. Perrin, P. I. Palmer, and P. F. Bernath, Atmos. Chem. Phys., 9, 3893-3910 (2009).

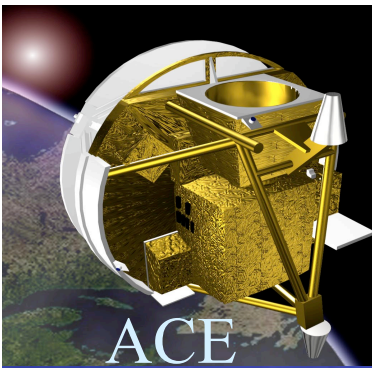


NO_x Descent



Ratio of NO_x relative to the average of years without enhancement (2005, 2007, and 2008)

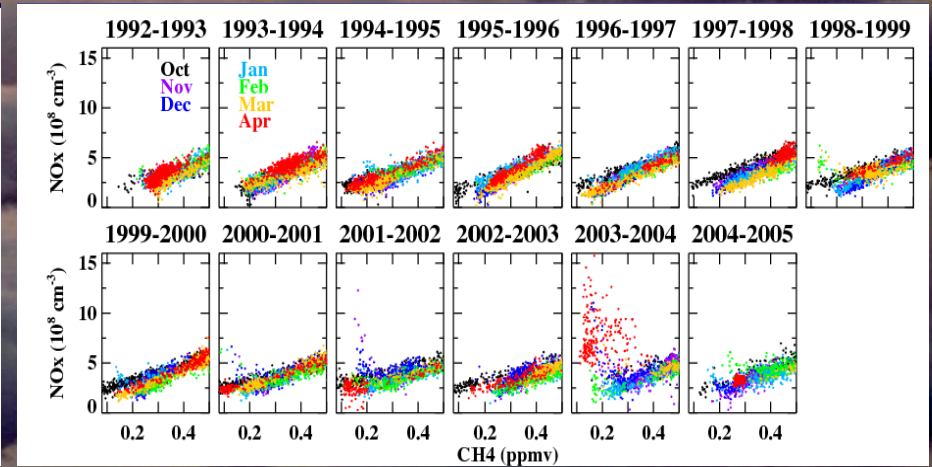
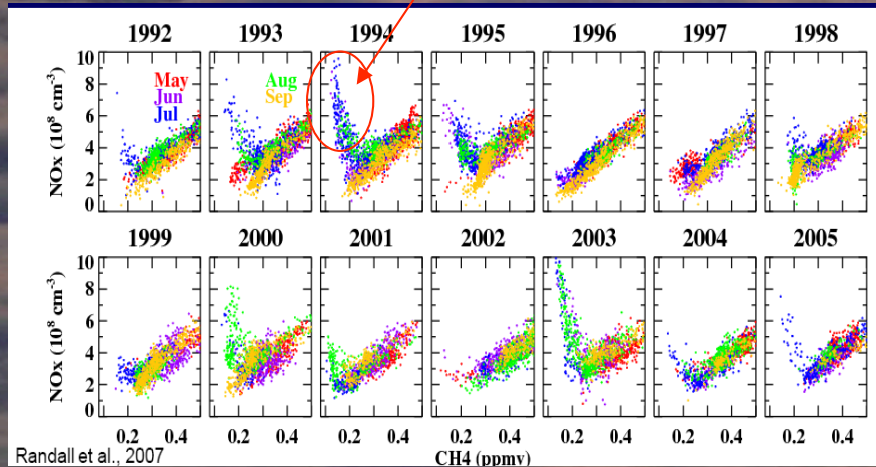
NO_x descent in the Arctic middle atmosphere in early 2009, C. E. Randall, V. L. Harvey, D. E. Siskind, J. France, P. F. Bernath, C. D. Boone, and K. A. Walker, *Geophys. Res. Lett.*, 36, L18811, doi:10.1029/2009GL039706 (2009)



All is quiet in the north, until...



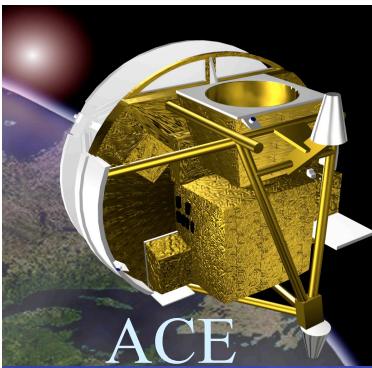
Enhanced NO_x created in the mesosphere descends into the stratosphere within the polar vortex



HALOE Southern Hemisphere

HALOE Northern Hemisphere

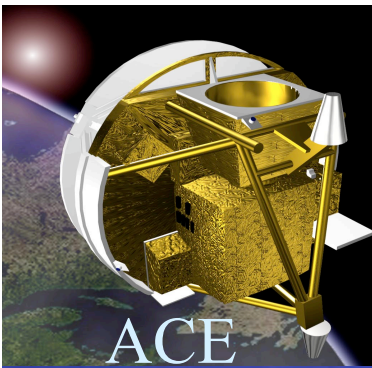
Energetic Particle Precipitation Effects on the Southern Hemisphere Stratosphere in 1992-2005, C.E. Randall, V.L. Harvey, C.S. Singleton, S.M. Bailey, P.F. Bernath, M. Codrescu, H. Nakajima and J.M. Russel III, *J. Geophys. Res.*, 112, D08308, doi:10.1029/2006JD007696 (2007)



New(?) threat to Arctic ozone



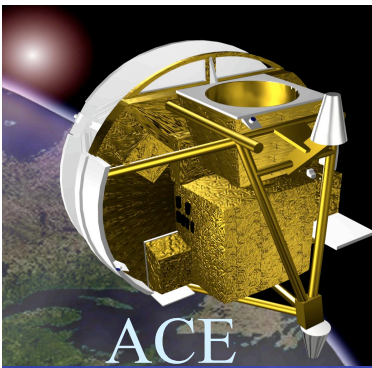
- How new is it? HALOE coverage extended into high latitudes in April. Would not see enhancements in February and March.
- In 2004, 2006, and 2009, observed descent of enhanced NO_x in the NH polar vortex.
- Originally attributed to strong energetic particle precipitation (EPP) from the solar storm of Halloween 2003, until ACE saw enhancements in 2006 and 2009 when there was weak EPP.



The new world order?



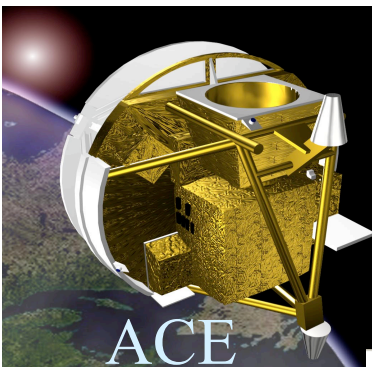
- Associated with a “remarkable” vortex recovery following a sudden stratospheric warming. A strong vortex led to enhanced adiabatic descent in the mesosphere.
- What was an unexpected occurrence in 2004 has now occurred 3 times in 6 years.
- Implications for Arctic ozone.
- Will the unusual meteorology linked to the effect persist? If so, what caused the change in atmospheric behavior?



It all seemed too easy...



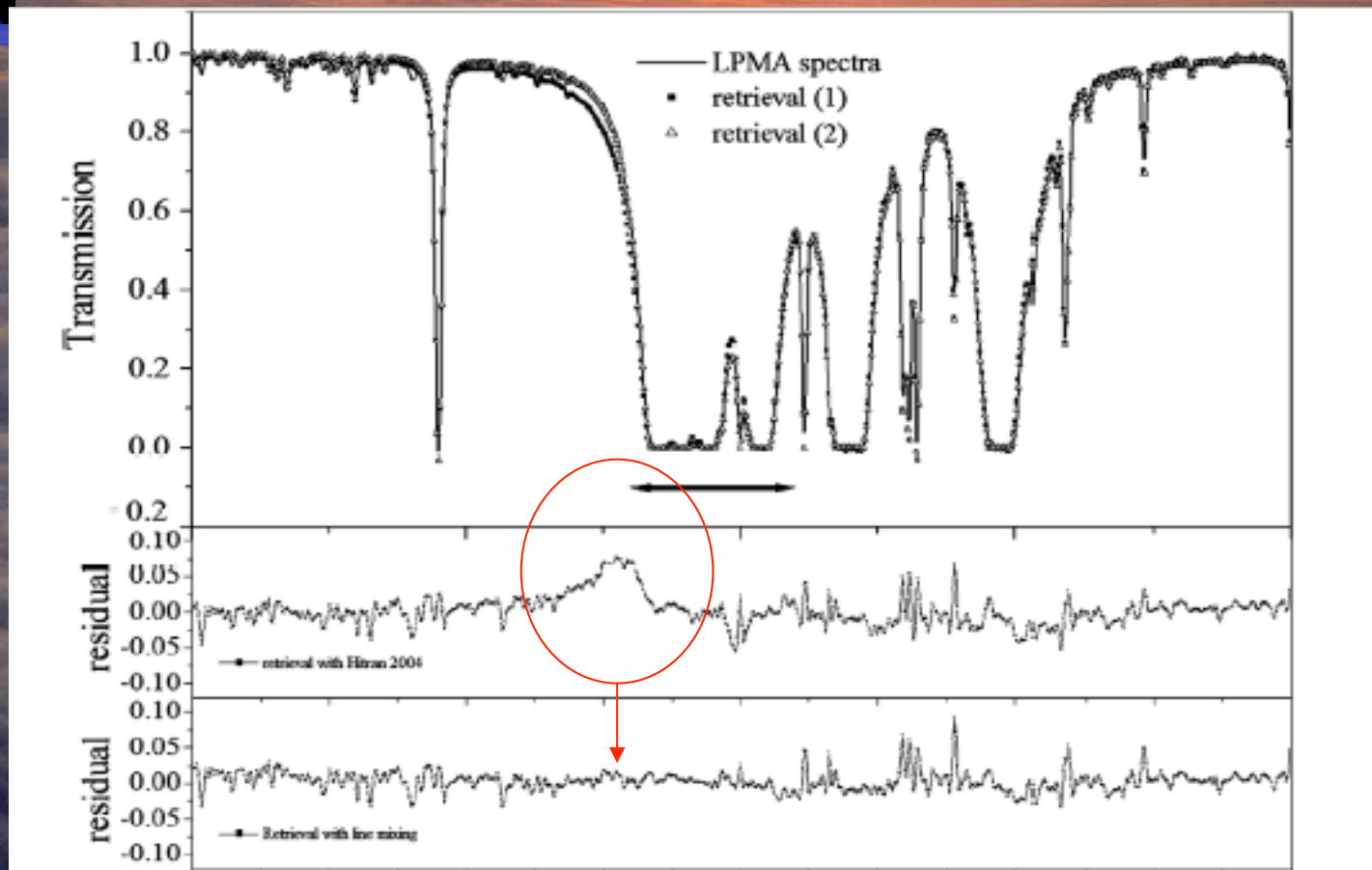
- Searching for weak spectral signatures in the troposphere (organics, CFCs, HCFCs).
- Bad residuals from stronger absorbers (e.g., CH_4 and H_2O) impeding the search.
- Limitations of using a simple Voigt.
- Implementing line mixing and alternative lineshapes (e.g., speed-dependent Voigt).
- Developed fast analytical approaches appropriate for “operational” retrievals.



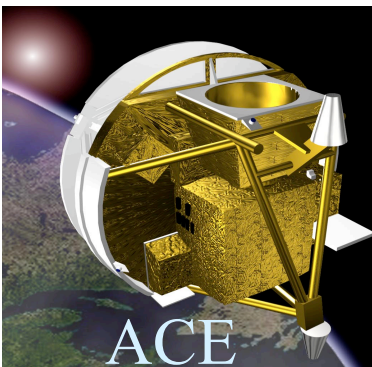
ACE



P9 manifold in the ν_3 band of CH_4



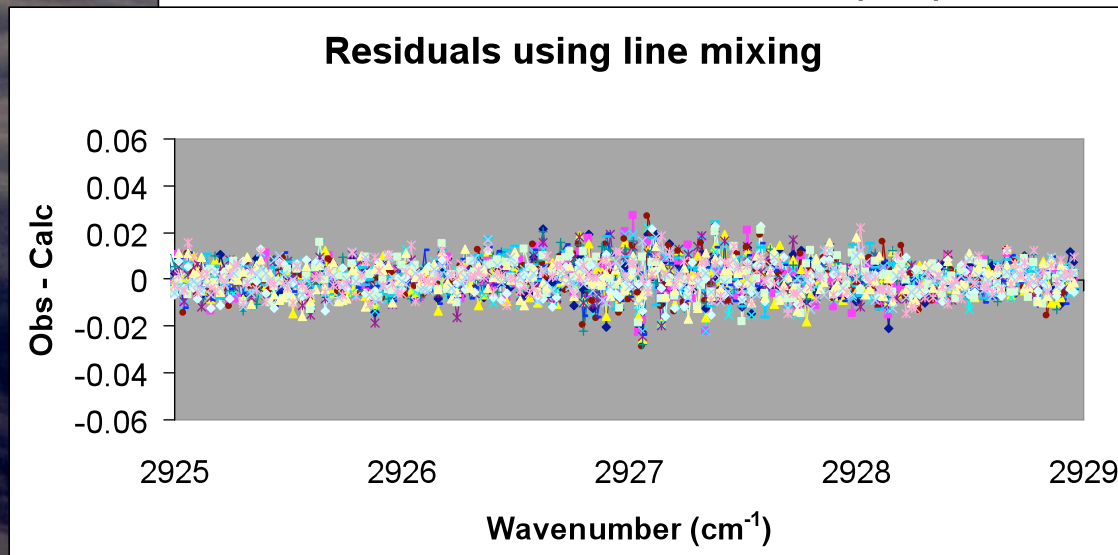
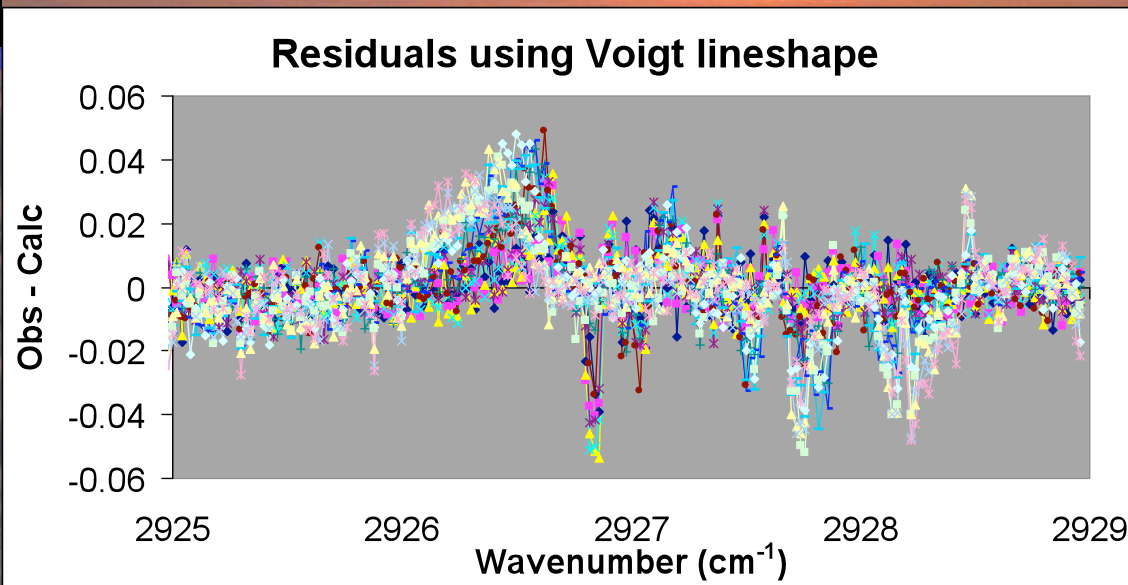
Measurement of the temperature dependence of line mixing and pressure broadening parameters between 296 and 90 K in the ν_3 band of 12CH_4 and their influence on atmospheric methane retrievals, Didier Mondelain, Sebastien Payan, Wenping Deng, Claude Camy-Peyret, Daniel Hurtmans, and Arlan Mantz, *J. Mol. Spec.*, 244, pp 130-137 (2007)



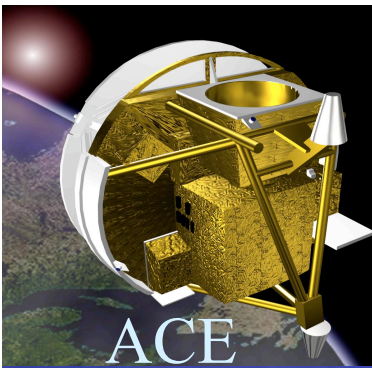
ACE



ACE results for P9



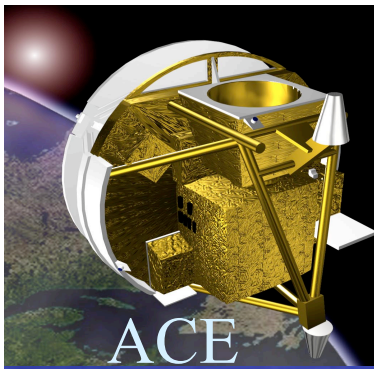
Along with line mixing parameters, also fitted intensities and the position for one of the lines.



Non-Voigt lineshape?



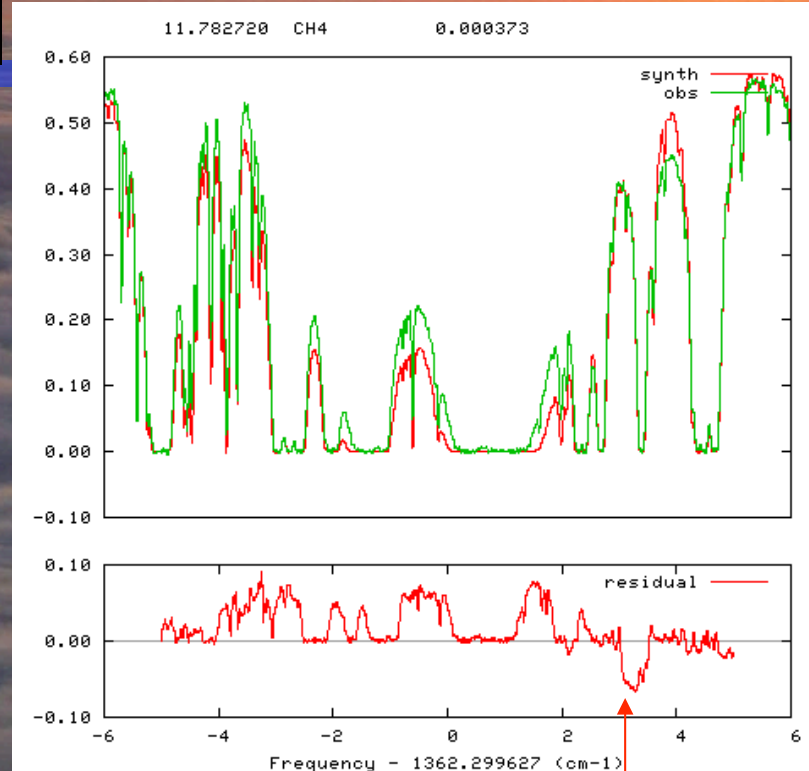
- Minor features remaining in the residuals.
- Could be a need for tweaked spectroscopic constants (width, T-dependence, position) for CH₄ lines or interferences.
- Could be deviations from Voigt lineshapes for the CH₄ lines. Residuals further reduced using speed-dependent Voigt lineshapes.
- Too much going on in atmospheric spectra. Need simpler CH₄ lab spectra to analyze. Plans to measure these spectra underway at York (U.K.).



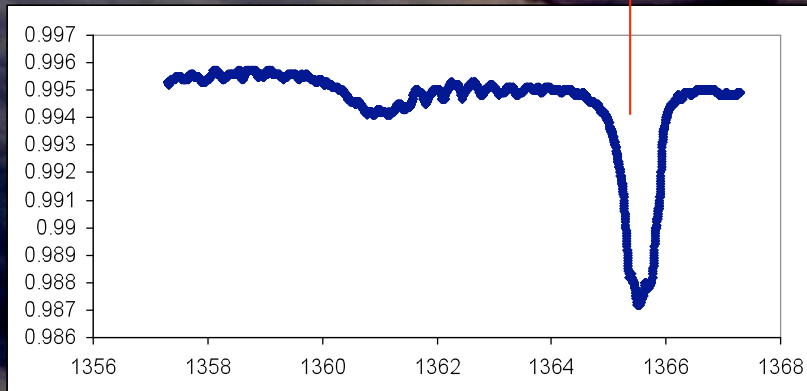
ACE

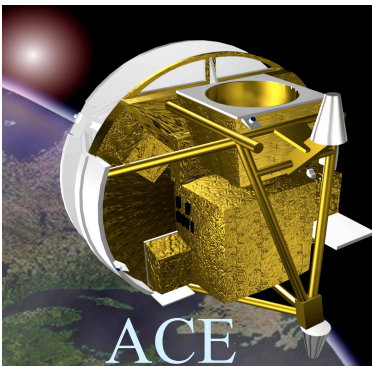


Acetone



Spectral signature for acetone visible in the residuals, but the region is polluted with bad residuals from both CH₄ (from line mixing) and H₂O.



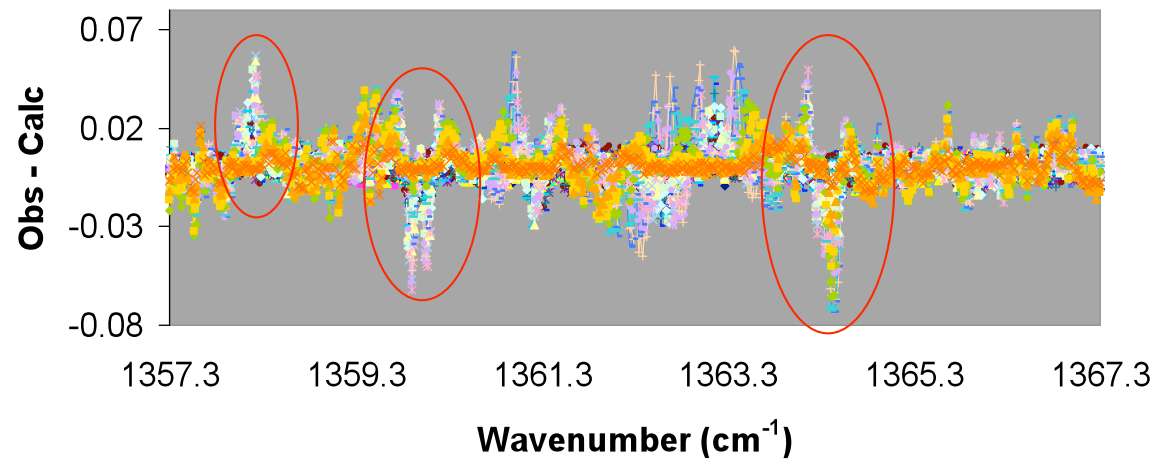


ACE

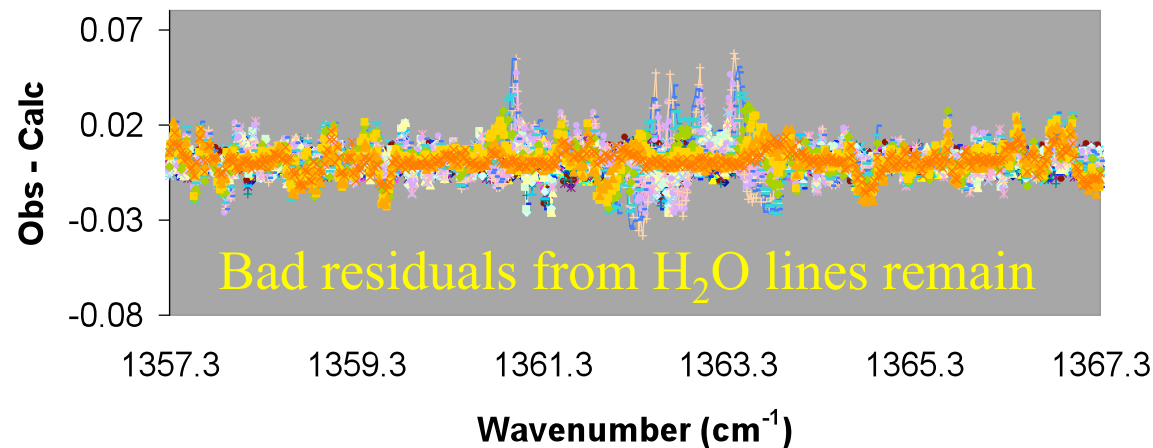


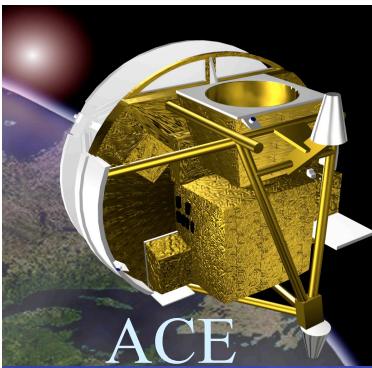
Add in line mixing

Residuals using Voigt lineshape



Residuals including line mixing in CH₄

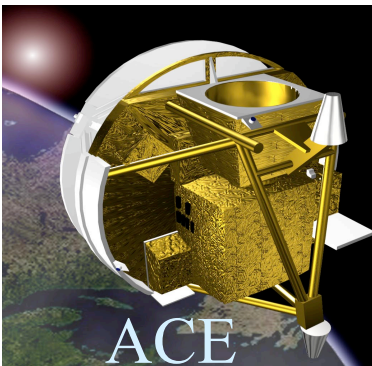




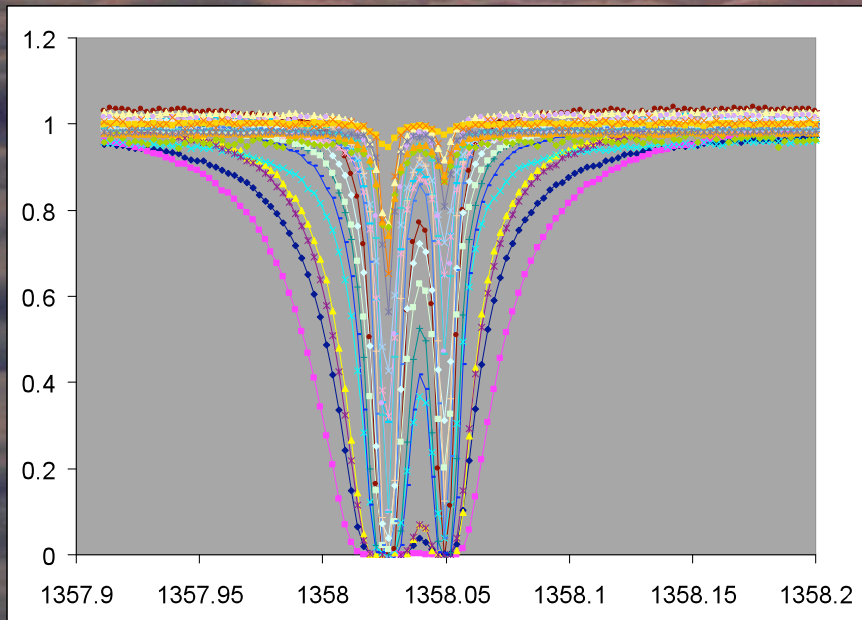
H₂O lineshape



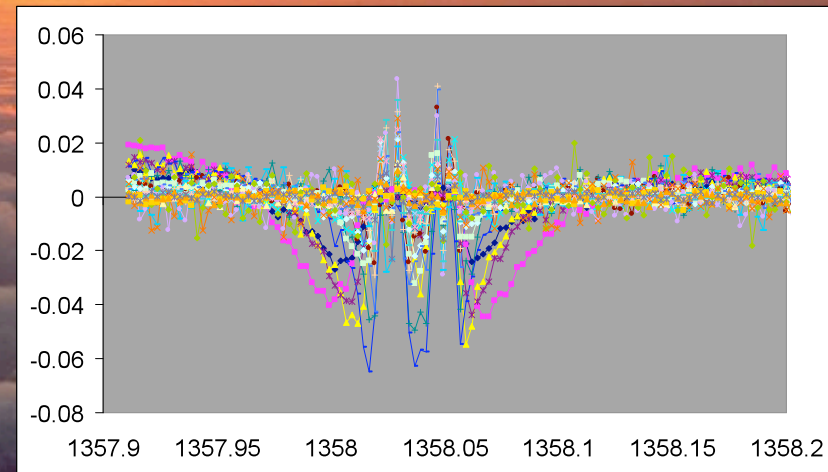
- Bad H₂O residuals cause problems for other molecules (e.g., acetone, HFC-134a,...).
- Not as straightforward as CH₄. Still haven't determined a reliable solution.
- Rapid change of H₂O VMR in troposphere a challenge for working on a 1-km grid.
- H₂O lab spectra from Manfred Birk at DLR.



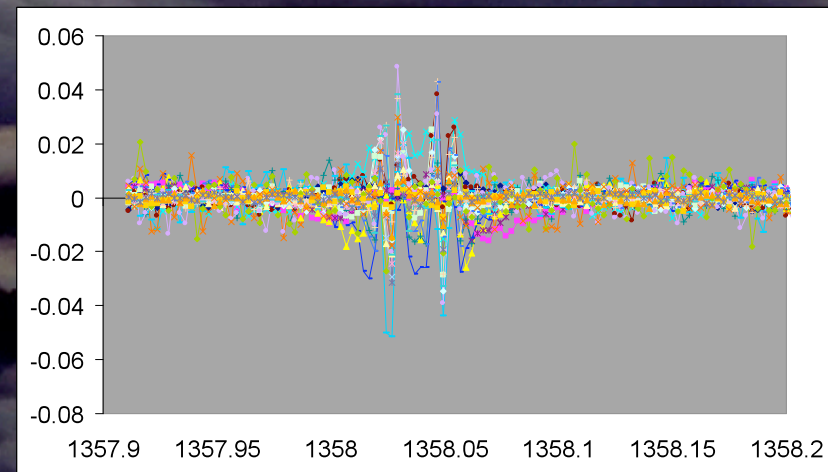
H₂O lab spectra



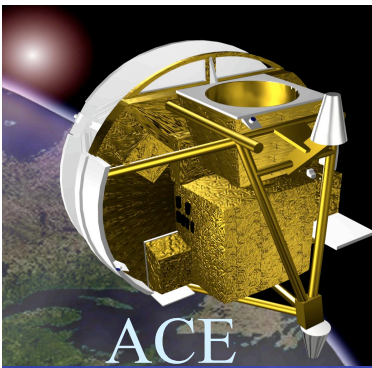
A set of 27 air broadened H₂O spectra, plus 4 self broadened



Residuals using Voigt function



Adjusting broadening parameters



Speed-dependence

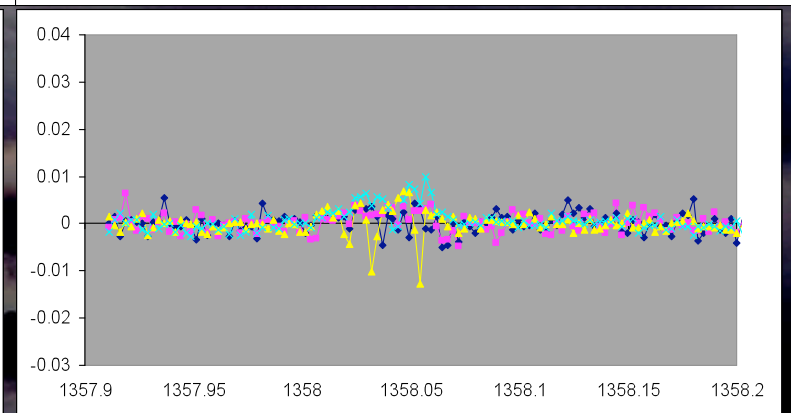
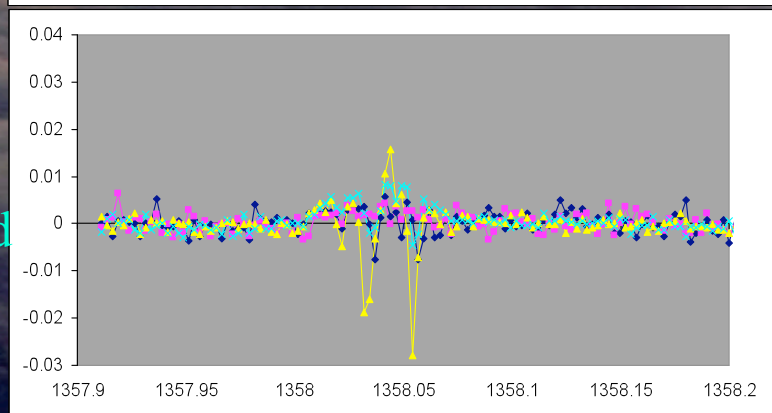
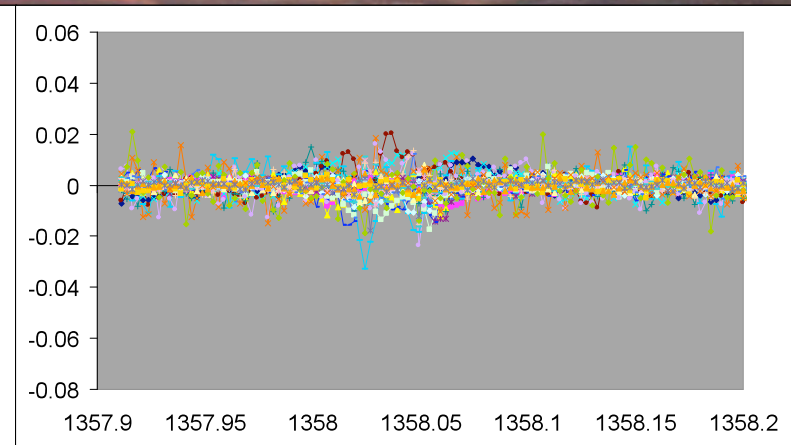
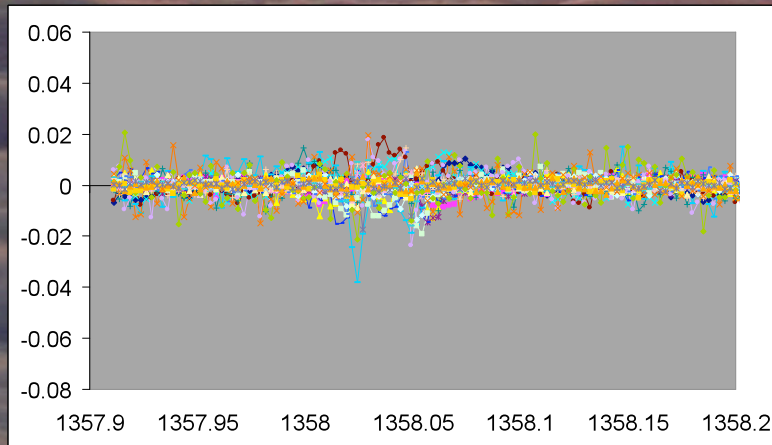


Air-broadened

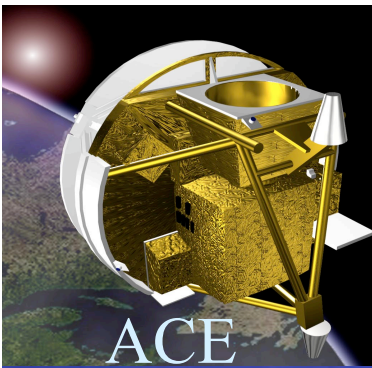
Self-broadened

$\eta = 0$

$\eta \neq 0$



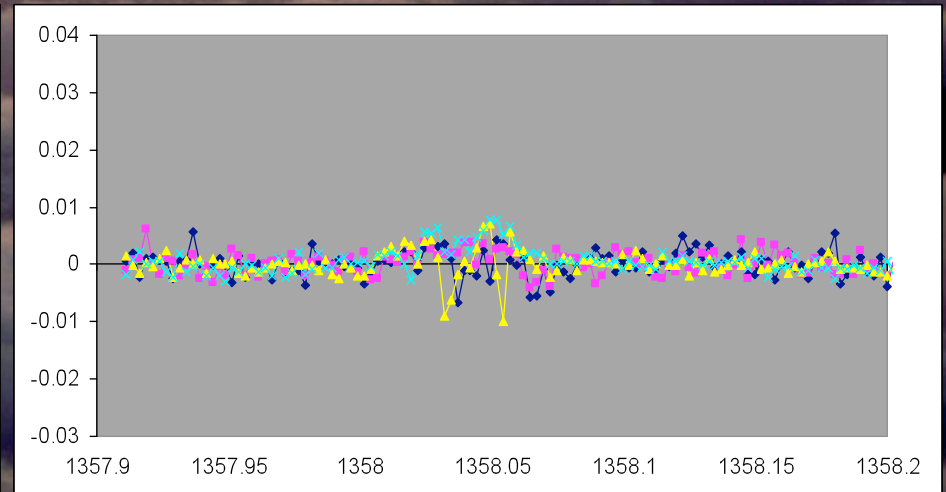
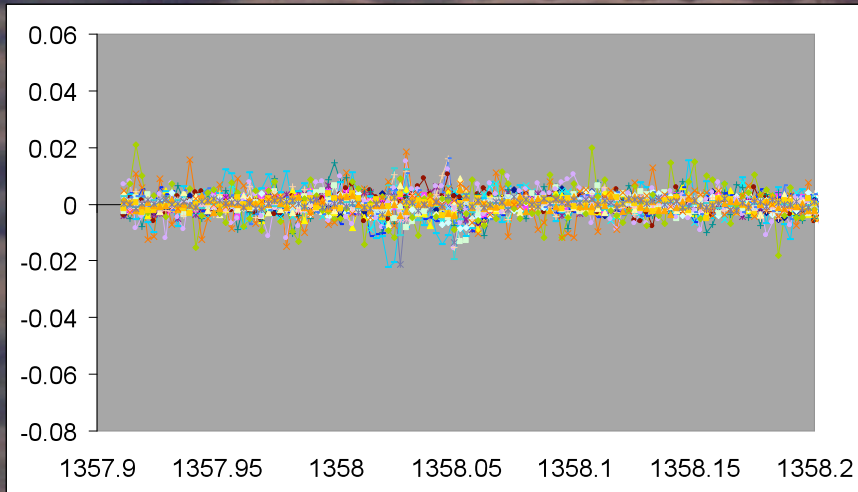
Speed-dependent Voigt profile for water vapor in infrared remote sensing applications, Chris D. Boone, Kaley A. Walker, and Peter F. Bernath, JQSRT, 105, 525-532 (2007).



A work in progress



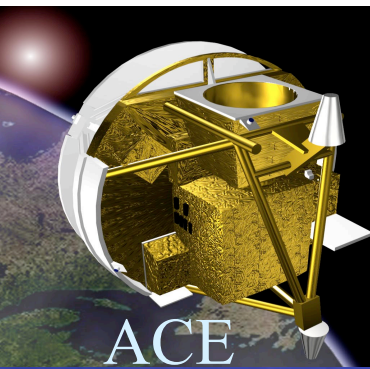
Separate T-dependence for self-broadening



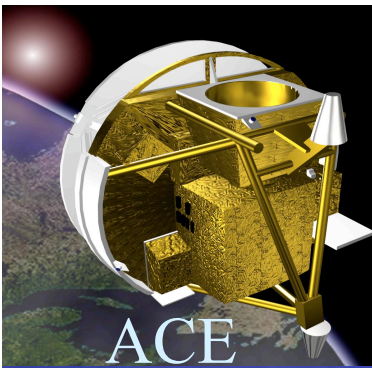
Note the different vertical scales. These residuals smaller than the air-broadened ones. Less noise.

Much work remains.

Summary



- ACE satellite continues to collect measurements.
- Start to look at trends.
- Further refining spectral calculations. Seeking weaker and weaker signatures.
- Generating spectroscopic parameters from lab spectra. Unavailable elsewhere.
- Version 2.2 data slated for public release.
- Version 3.0 underway. A few months to catch up.
- Follow-on mission? Some preliminary studies.



Acknowledgement



Thanks to the Canadian Space Agency for project funding.