ATMOS on ISS

Proposed Deployment of the Atmospheric Trace Molecule Spectroscopy Experiment on the International Space Station

Scientific Objectives

November 20, 1995

This document is a draft of a white paper providing scientific justification for deployment of the ATMOS instrument on the International Space Station. It is distributed to solicit feedback from the scientific community. Any feedback you can provide on this manuscript is appreciated.

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Executive Summary

The ATMOS Fourier Transform Spectrometer is an extant instrument which acquires high-resolution, broadband solar occultation spectra from space. ATMOS uses infrared transmission to profile over 30 atmospheric constituents including O_3, N_2O, CH_4, H_2O, the entire NO_y family, and much of the chorine and fluorine families, including HCl, HF, and $ClONO_2$. ATMOS has operated successfully in four Space Shuttle missions and in an extensive series of ground-based measurements. Deployment on the International Space Station (ISS) will increase by orders of magnitude the number of observation opportunities, providing greater latitudinal, seasonal, and temporal coverage than achieved previously, and will extend the ATMOS database from 1985 into the next two decades.

Key science objectives of an ATMOS deployment on ISS are:

- Transport studies; capitalizing on the ability to make highly precise measurements of key tracers of atmospheric transport through different seasons and over several years.
- Stratospheric chemistry; providing measurements of gases important for the photochemical regulation of O_3 over a wide range of conditions, taking full advantage of co-located measurements of aerosols by SAGE III.
- Tropospheric chemistry; using high quality tropospheric spectra to measure concentrations of H_2O, O_3, CO, HNO_3, and other species important for regulating OH.
- Polar processes; providing observations of ozone, nitrogen oxides, water, inorganic chlorine, and long-lived gases during periods of vortex breakup.
- Radiation and climate; recording changes in solar infrared transmission, as a result of changes in aerosols and tropospheric water vapor, which directly impact climate.
- Long-term monitoring; building on the ATMOS data record to measure growth rates of greenhouse gases and precursors of O_3 depleting catalysts.
- New species detection; fingerprinting the state of the atmosphere with high quality infrared spectra to preserve a record of gases which are growing in importance.
- Correlative measurements; providing a reference standard for comparison with the suite of measurements from EOS Chem-1, bridging the gap between UARS and EOS.

ATMOS's previous accomplishments demonstrate these science objectives are realistic. A deployment on International Space Station will contribute much to the goals of the Mission to Planet Earth Program.

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Background

The ATMOS (Atmospheric Trace Molecule Spectroscopy) Fourier Transform Spectrometer is an extant instrument (**Fig. 1**) with a proven record of profiling over 30 atmospheric constituents from space. ATMOS is a Michelson interferometer characterized by high spectral resolution (0.01 cm^{-1}) coupled with a broadband spectral response (600 to 4800 cm^{-1} or 2 to 16 μm). The instrument is optimized to operate with a rapid scan time to facilitate space-based solar occultation measurements. Constituent profiles are inferred from atmospheric transmission spectra, which provide a spectroscopic fingerprint of the atmosphere (**Fig. 2**).

ATMOS is able to make accurate and precise measurements of concentration profiles of a large number of atmospheric constituents, including several isotopomers of the more abundant gases. Species presently retrieved on a routine basis (**Fig. 3**) include O_3, N_2O, CH_4, H_2O HCl, HF, HNO_3, NO, NO_2, $ClONO_2$, N_2O_5, HNO_4, CFC-11 (CCl_3F), CFC-12 (CCl_2F_2), HCFC-22 (CHF_2Cl), SF_6, OCS, HCN, HDO, $H_2{}^{17}O$, $H_2{}^{18}O$, CH_3D, CO, CCl_4, C_2H_2, C_2H_6, CH_3Cl, and CF_4. Other gases whose retrieval from ATMOS spectra have been demonstrated include COF_2 and SO_2.

ATMOS spectra are self-calibrated and insensitive to long term drifts in instrument performance. The accuracies of measured concentration profiles are limited primarily by the quality of the infrared spectroscopic line parameters or cross-sections, which are continually improved through laboratory measurements. Many atmospheric gases, such as N_2O_5 and HNO_4, have broadband absorption features and can only be measured using an instrument with high spectral resolution and broadband response, as the fine absorption structure of interfering gases must be distinguished from the broadband features of the target gas. The resolution and broad response of ATMOS also renders the retrievals insensitive to changes in background transmission, as might be encountered following major volcanic eruptions leading to heavy aerosol loading in the atmosphere.

The ATMOS instrument has proven its ability to operate reliably from space during four Space Shuttle missions (**Fig. 4**). The first deployment of ATMOS was part of the Spacelab 3 payload aboard the Space Shuttle *Challenger*, launched on 29 April 1985. Subsequent ATMOS deployments, as part of the ATLAS Space Shuttle payloads occurred in March 1992, April 1993, and November 1994. ATMOS measurements from these missions have led to over 80 publications in the peer-reviewed literature, which have focused on photochemical studies of nitrogen and chlorine bearing gases that regulate ozone, solar science, trends in chlorine and fluorine loading related to anthropogenic activity, and the chemistry and dynamics of the polar vortices following the rapid spring-time removal of ozone.

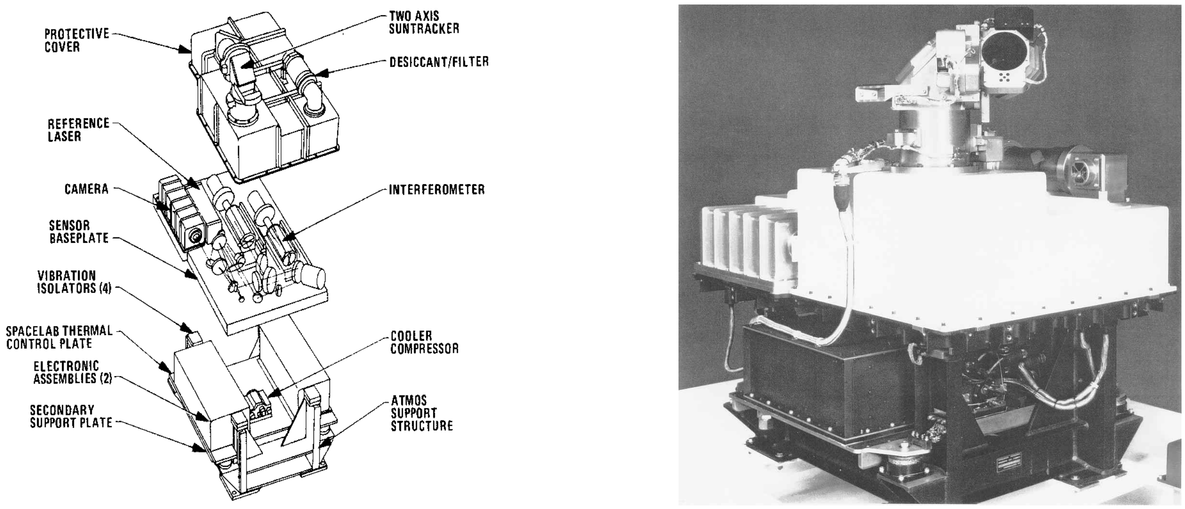

Figure 1: The ATMOS Fourier Transform Interferometer. The schematic on the left illustrates the optics and electronic subsystems; the assembled instrument is shown on the right.

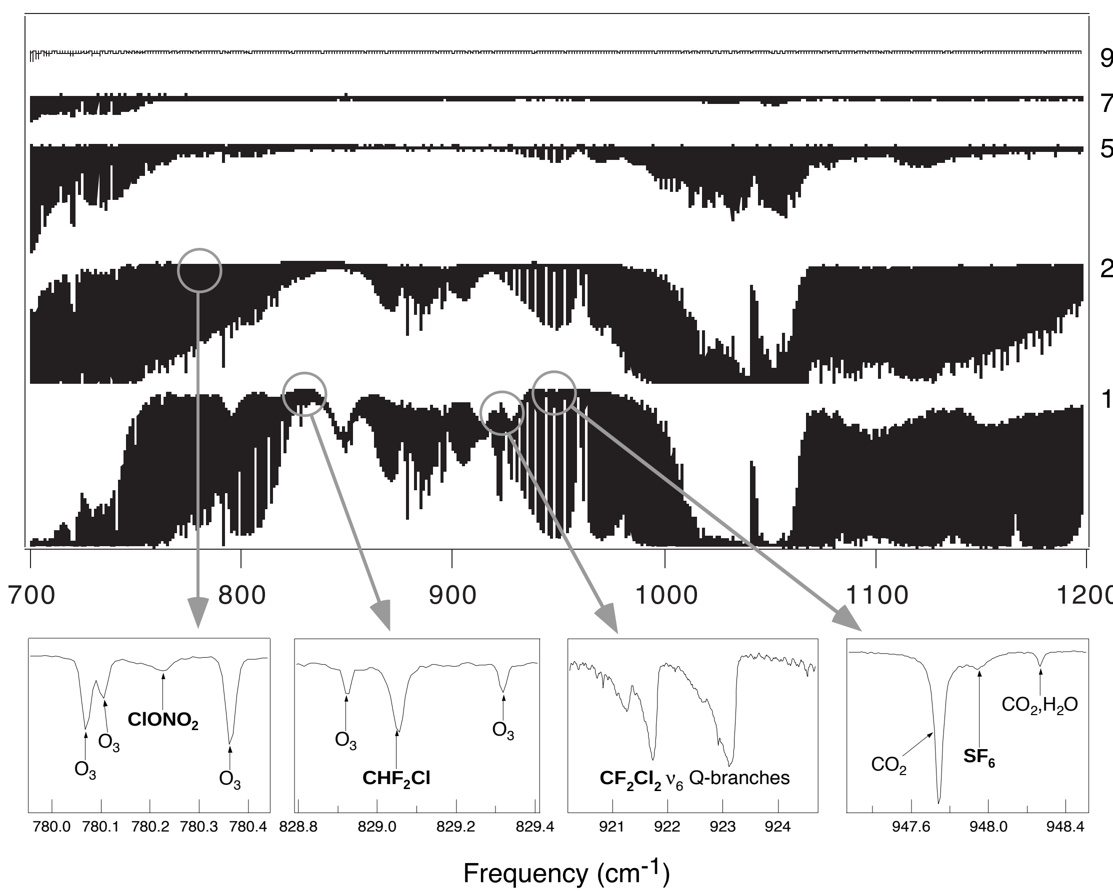

Figure 2: Typical ATMOS spectra. The upper figure shows the same broad spectral region in successively lower altitude observations. These spectra have been offset in the vertical axis for clarity. The lower figures illustrate smaller spectral intervals used for trace gas retrievals. Vertical scales differ among displayed spectra.

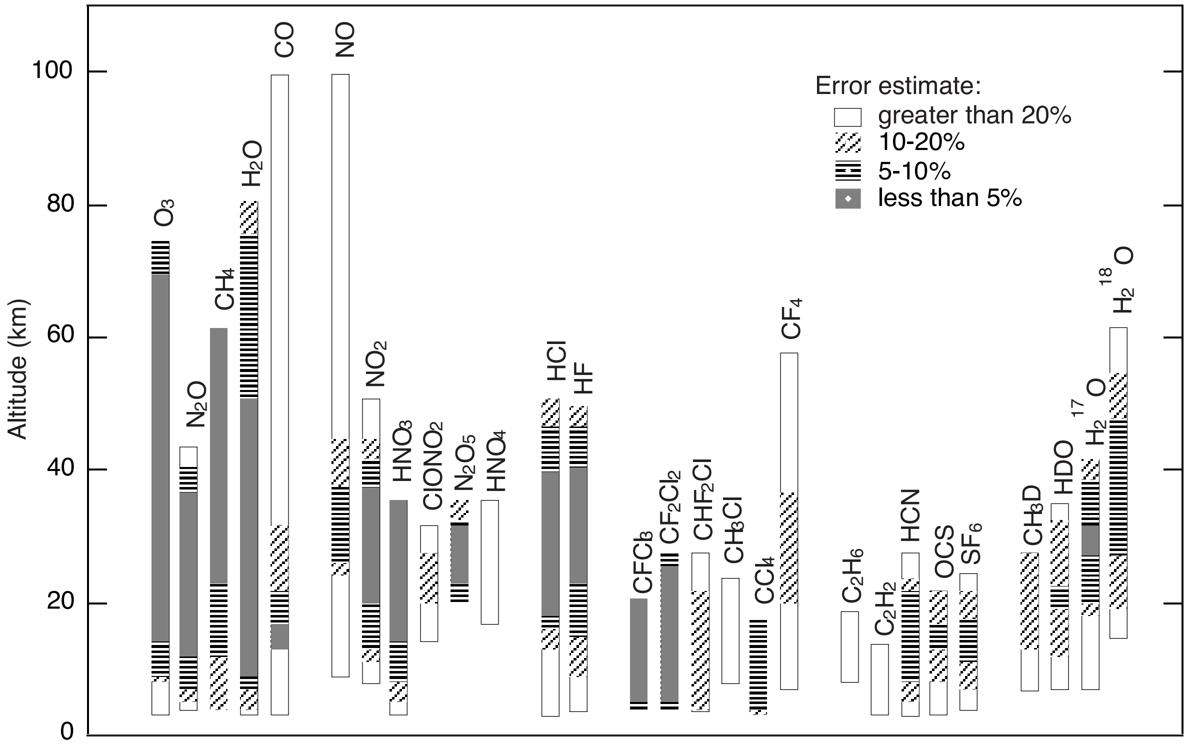

Figure 3: Approximate vertical range and random errors for routinely retrieved species. Errors represent those associated with ATMOS version 2 data.

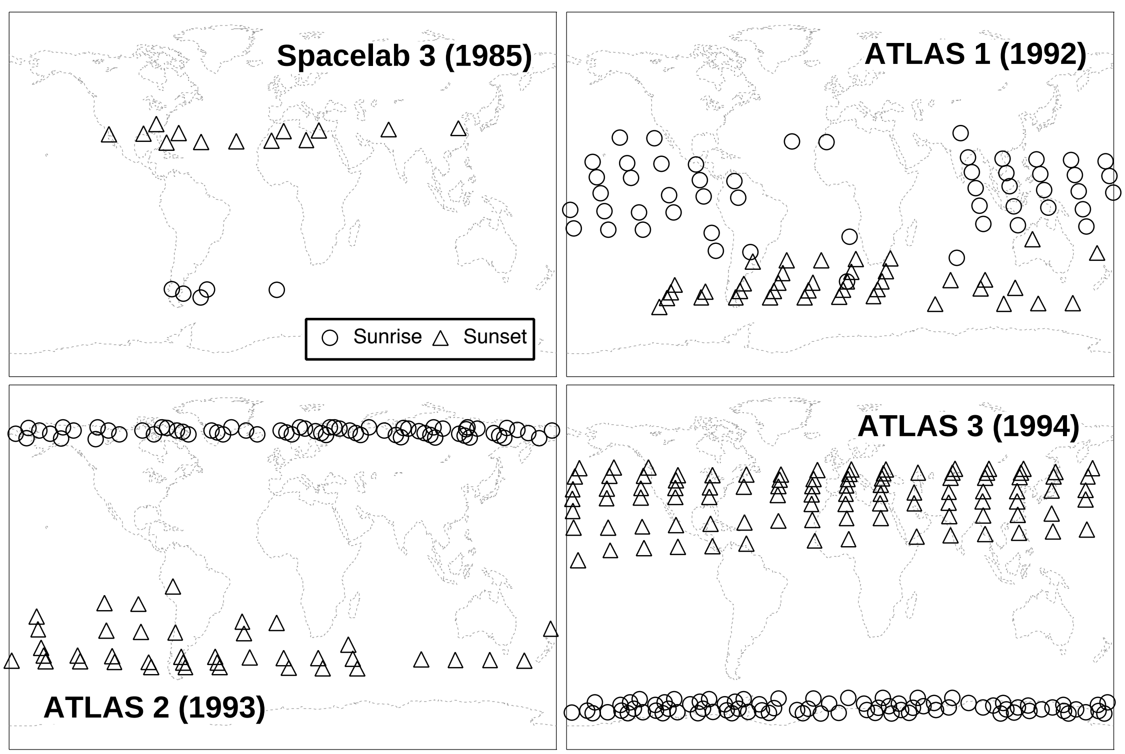

Figure 4: Map showing the distribution of ATMOS observations during each of four Space Shuttle missions. Spacelab 3: April 29-May 7, 1985; ATLAS-1: March 24-April 3, 1992; ATLAS-2: April 8-16, 1993; ATLAS-3: November 3-14, 1994.

The International Space Station (ISS) presents an ideal platform for continued operation of ATMOS. Deployed in stages, ISS is expected to be fully operational in mid-2002 and to have a life of at least 10 years. ATMOS has been identified as a candidate to fill a berth on an Express Pallet to be mounted on the ISS in February 2001 (**Fig. 5**). The 51.6° inclination orbit of the ISS allows access to ~12,000 solar occultations per year, providing coverage from 45° S to 45° N in all seasons, extending to 70° S or 70° N in summer (**Fig. 6**). Deployment of ATMOS on ISS will provide an unprecedented data set, complementing the intensive measurements from EOS Chem-1, other space-borne platforms, as well as UAVs (unmanned airborne vehicles), aircraft, balloons and ground-based instruments.

Synergies with Planned Programs

ATMOS has demonstrated concentration measurements of sufficient accuracy and resolution to complement the high precision *in situ* data acquired by instruments aboard research aircraft such as the NASA ER-2 (**Fig. 7**). ATMOS measurements provide relations of tracers of atmospheric transport and distributions of reactive nitrogen and chlorine gases to higher altitudes and over broader geographic regions than possible with *in situ* data. Deployment of ATMOS on ISS will provide a baseline of measurements bridging the multi-year gap between the UARS and EOS Chem-1 programs. Continued observations by ATMOS will provide a transfer standard by which future measurements can be referenced to existing observations.

The next generation of SAGE (Stratospheric Aerosols and Gas Experiment) instruments, one of which is planned for deployment on ISS, provides a particularly powerful capability in tandem with ATMOS. SAGE III features an 800 channel visible spectrometer (280 to 1030 nm) to measure O_3, H_2O, NO_2, and aerosols in solar occultation mode (NO_3 and OClO are observed in lunar mode). Together ATMOS and SAGE III will provide spectral measurements covering the IR and UV-VIS portions of the spectrum, and will monitor identical air masses. The large number of trace gases measured by ATMOS combined with the aerosol measurements of SAGE III will provide a unique data set for studying atmospheric photochemical and dynamical processes as well as climate.

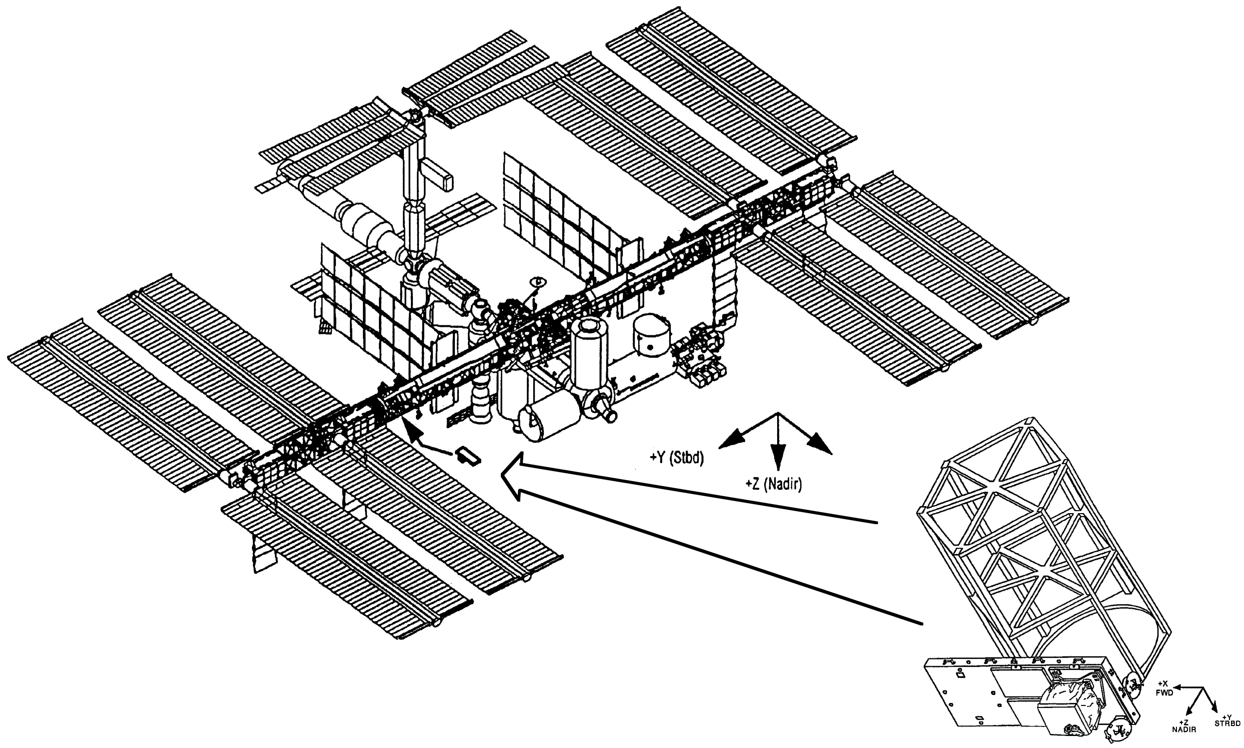

Figure 5: Proposed configuration of ATMOS on International Space Station. The ATMOS instrument will be attached to an ISS Express Pallet which will be located on the ISS structure as shown.

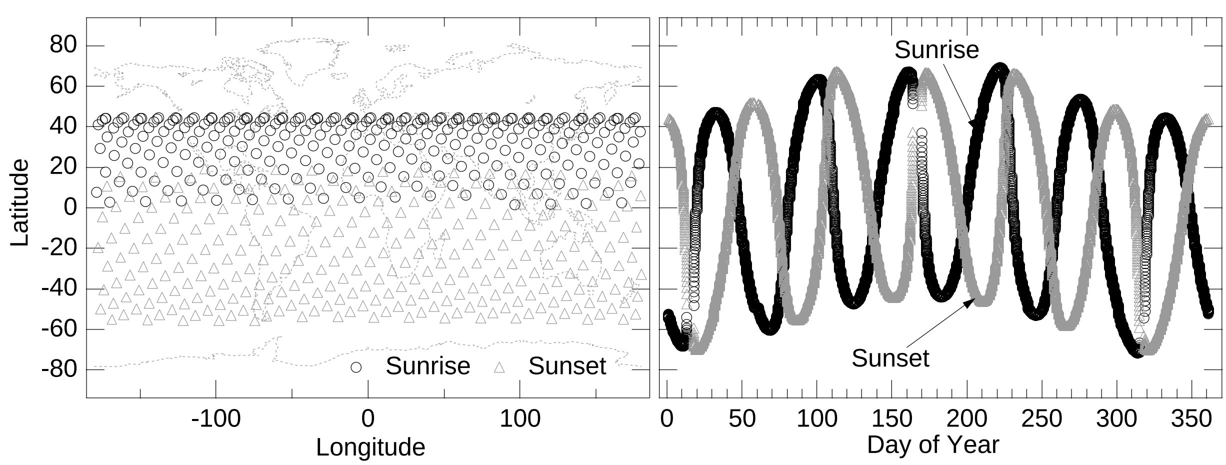

Figure 6: Distribution of solar occultations from the orbit of ISS. The left panel shows the latitude and longitude of available sunset and sunrise observations over an 18 day period in early December. The right panel shows the latitude of available observations over the course of a year.

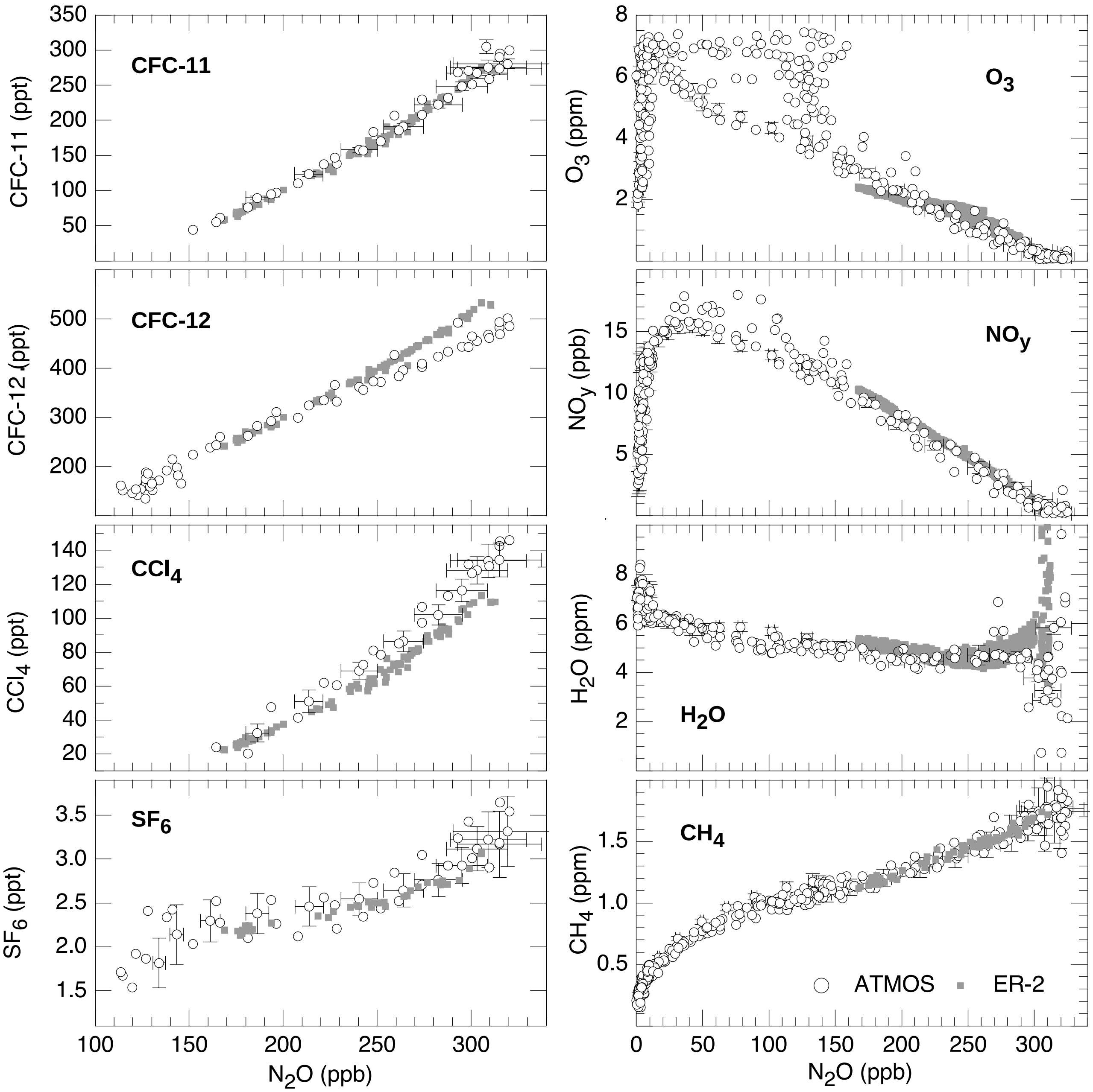

Figure 7: Comparison of ATMOS/ATLAS-3 and ER-2 ASHOE/MAESA aircraft measurements at northern mid-latitudes during early November 1994. ATMOS data extend the *in situ* relations measured from aircraft. Differences between ATMOS and *in situ* measurements of CFC-12 and CCl_4 are currently under investigation. ER-2 measurements courtesy of M. Loewenstein (N_2O), J. W. Elkins (CFC-11, CFC-12, CCl_4, SF_6, and CH_4), M. H. Proffitt (O_3), D. W. Fahey (NO_y), and K. K. Kelly (H_2O).

Science Objectives

The deployment of ATMOS on ISS will provide measurements which will improve our understanding of transport and chemical processes in the stratosphere and troposphere, and establish a fundamental record of long-term changes in atmospheric composition and infrared transmission, due to both natural and anthropogenic sources.

Transport

A deployment on ISS will particularly enhance the applicability of ATMOS data for examining atmospheric transport. Air enters the stratosphere mainly at the tropical tropopause and is lofted vertically, eventually descending at mid-latitudes. The upwelling of tropical air is accompanied by both detrainment air to mid-latitudes and entrainment of mid-latitude air. These rates of exchange are not well constrained by current observations. ATMOS measures with a high degree of precision the distributions of a large number of long-lived tracers of atmospheric transport, each of which has a different photochemical lifetime (e.g., O_3, NO_y, CH_4, N_2O, CFCs, SF_6, HF, CO). The distributions of gases and their ratios (e.g., NO_y/O_3, NO_y/N_2O) are sensitive measures of efficiencies of transport, serving to identify barriers to transport between different geographic regions and the variation and extent of these barriers with season and phase of the quasi-biennial oscillation (QBO) of tropical winds. ATMOS measurements of tracer-tracer correlations will be used, for example, to evaluate rates of entrainment and detrainment between the tropics and mid-latitudes as a function of altitude. Folds and structures seen by ATMOS in vertical profiles and tracer-tracer correlations of gases such as N_2O, NO_y, and CH_4 will permit the examination of large-scale mixing events between air from different geographical regions (c.f. **Fig. 8**).

Deployment of ATMOS on ISS will provide tracer measurements with complete seasonal and latitudinal coverage over the tropics, mid, and high-latitudes. Full longitudinal coverage will be obtained every 24 hours (16 sunrise and 16 sunsets), and full latitudinal coverage for both sunrises and sunsets will be obtained in a one month observation period (the density of measurements will be similar to that of the Halogen Occultation Experiment on UARS). Maps of tracer distributions with altitude, latitude, longitude, and season (c.f. **Fig. 9**) will provide an unprecedented opportunity to measure parameters that govern mixing and transport on a global scale, providing essential constraints for 2D and 3D models used to simulate distributions of ozone.

The global loss rate of atmospheric gases whose primary sinks are in the stratosphere (e.g., N_2O, CH_4, CFCs) is dominated by loss in the tropics, due to the high intensity of solar radiation and the upward lofting of air to regions of increasing photochemical activity. Currently, there are few measurements of the tropical profiles of many of these gases,

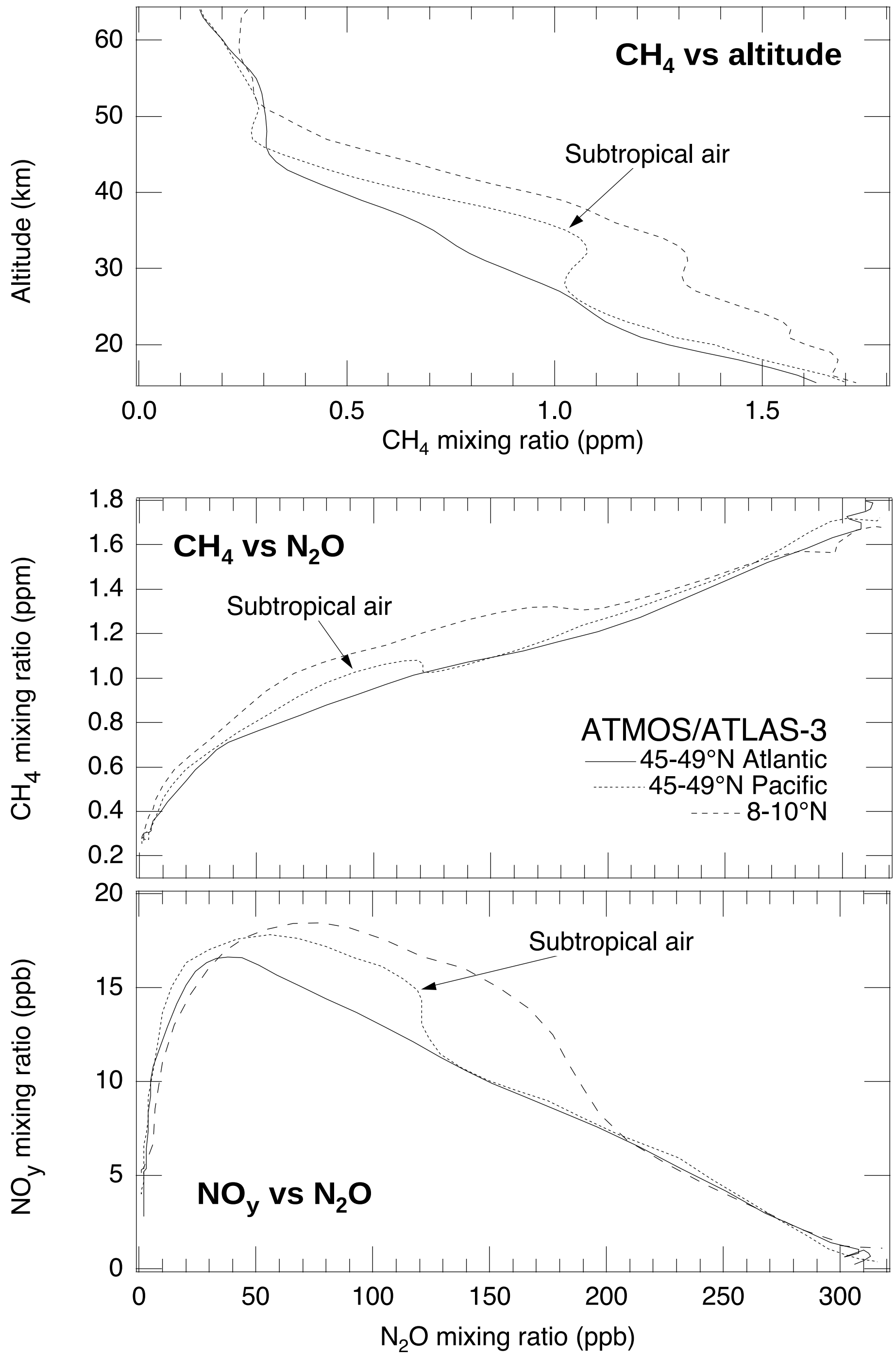

Figure 8. Tropical and northern mid-latitude CH_4, NO_y and N_2O from ATMOS/ATLAS-3 during early November, 1994. The increase of CH_4 and NO_y over the Pacific Ocean in northern mid-latitudes compared to that over the Atlantic indicates transport from the subtropics at mid-stratospheric altitudes.

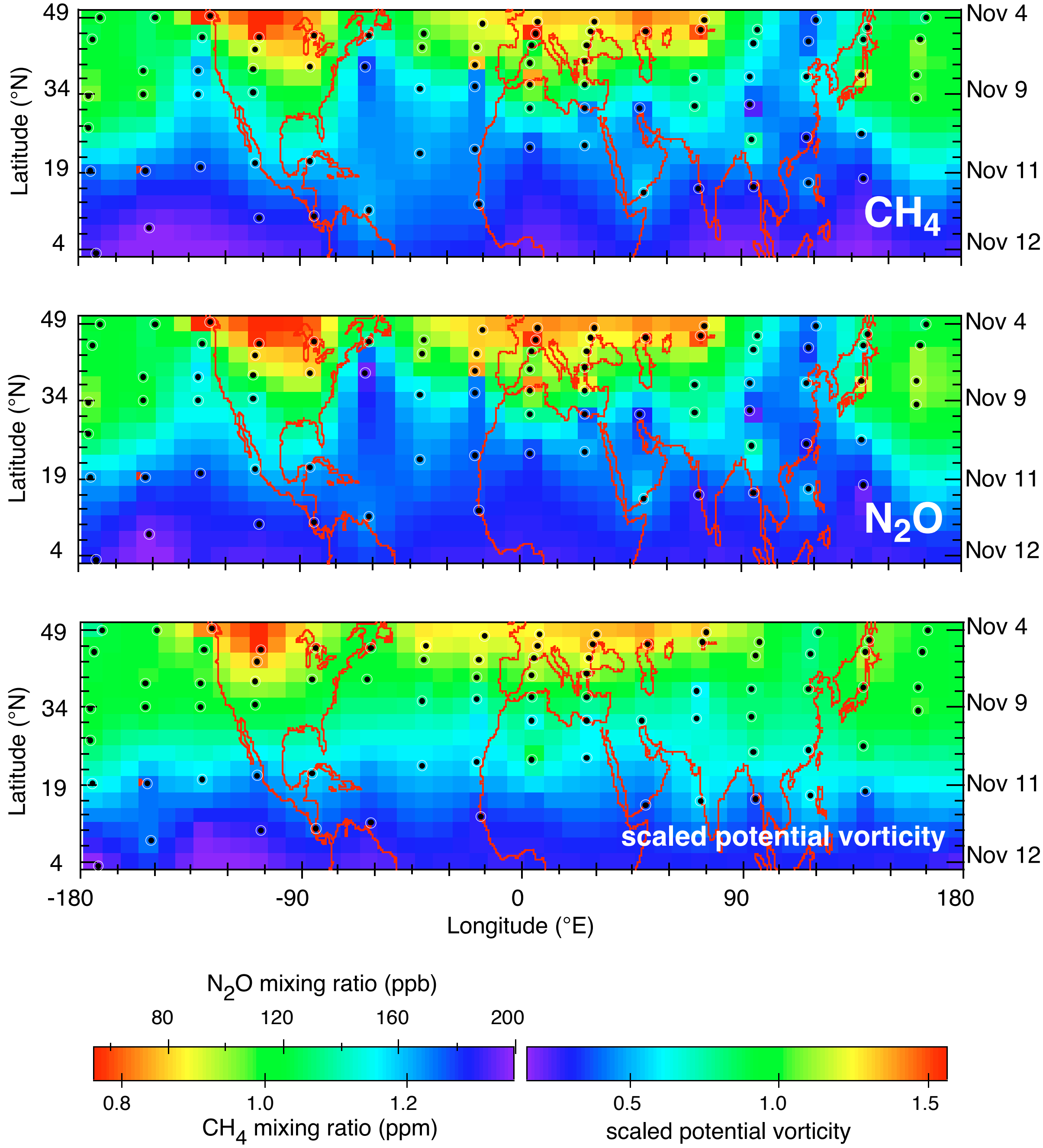

Figure 9: Northern mid-latitude N_2O, CH_4, and scaled potential vorticity (sPV) maps on the 750 K potential temperature surface (approximately 13 mbar or 29 km) from ATMOS on ATLAS-3, November 1994. The open circles on the maps indicate measurement locations. The density of coverage from an ISS deployment would be similar, with more complete latitude coverage and less gaps in observations due to interferences. The sPV map was generated from U. K. Meteorological Office data (thanks to R. Swinbank and A. O'Neill).

especially over a wide range of seasons. Accurate measurements by ATMOS of the distribution of trace gases, particularly in the tropics, will lead to an evaluation of their total atmospheric burdens and integrated photochemical lifetimes.

Measurement by ATMOS of total hydrogen (H_2O+2CH_4) will record the vertical propagation of the seasonal variation of water entering the stratosphere, thus providing a means to directly estimate vertical velocities in the tropics, and their change with season (**c. f. Fig. 10**). Finally, ATMOS measurements of the isotopes HDO and CH_3D can be used to diagnose stratospheric/tropospheric exchange processes, as the vertical variation of stratospheric enrichment ratios in conjunction with kinetic modeling provides information on the temporal history of gas parcels originating from the troposphere.

Ko, M. K. W., N. D. Sze, and D. K. Weisenstein, The roles of dynamical and chemical processes in determining the stratospheric concentration of ozone in one-dimensional and two-dimensional models, *J. Geophys. Res.*, *94*, 9889-9896, 1989.

Mote, P. W., K. H. Rosenlof, J. R. Holton, R. S. Harwood, and J. W. Waters, Seasonal-variations of water-vapor in the tropical lower stratosphere, *Geophys. Res. Lett.*, *22*, 1093-1096, 1995.

Murphy, D. M., D. W. Fahey, M. H. Proffitt, S. C. Liu, K. R. Chan, C. S. Eubank, S. R. Kawa, and K. K. Kelly, "Reactive nitrogen and its correlation with ozone in the lower stratosphere and upper troposphere, *J. Geophys. Res.*, *98*, 8751-8773, 1993.

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Stratospheric Chemistry

Understanding the role of a variety of photochemical processes that regulate stratospheric O_3 requires simultaneous measurement of reactive radical constituents, reservoir species, and long-lived precursors of radicals. ATMOS measures vertical profiles of nearly every species of the nitrogen oxide family (NO_y: NO, NO_2, N_2O_5, HNO_4, HNO_3, and $ClONO_2$), in addition to important precursors to radical concentrations (O_3, N_2O, CFCs, H_2O, and CH_4). ATMOS also measures all the major components of the chlorine and fluorine families (HF, HCl, $ClONO_2$, COF_2, CFC-11, CFC-12, HCFC-22, CCl_4, CH_3Cl, CF_4 and SF_6). The co-manifested SAGE III instrument will provide simultaneous measurements of aerosol surface area, essential for quantifying the rates of heterogeneous reactions. These measurements are sufficient to constrain many of the important photochemical reactions regulating O_3. Together with profiles of OH and ClO from MLS (Microwave Limb Sounder) on EOS Chem-1, they represent an unprecedented capability.

Data from the past ATMOS missions have provided the best available constraints for understanding processes that regulate the abundance of nitrogen radicals and chlorine reservoirs over broad ranges of altitude and latitude. ATMOS measurements were important for elucidating the role of heterogeneous processes occurring on sulfate aerosols,

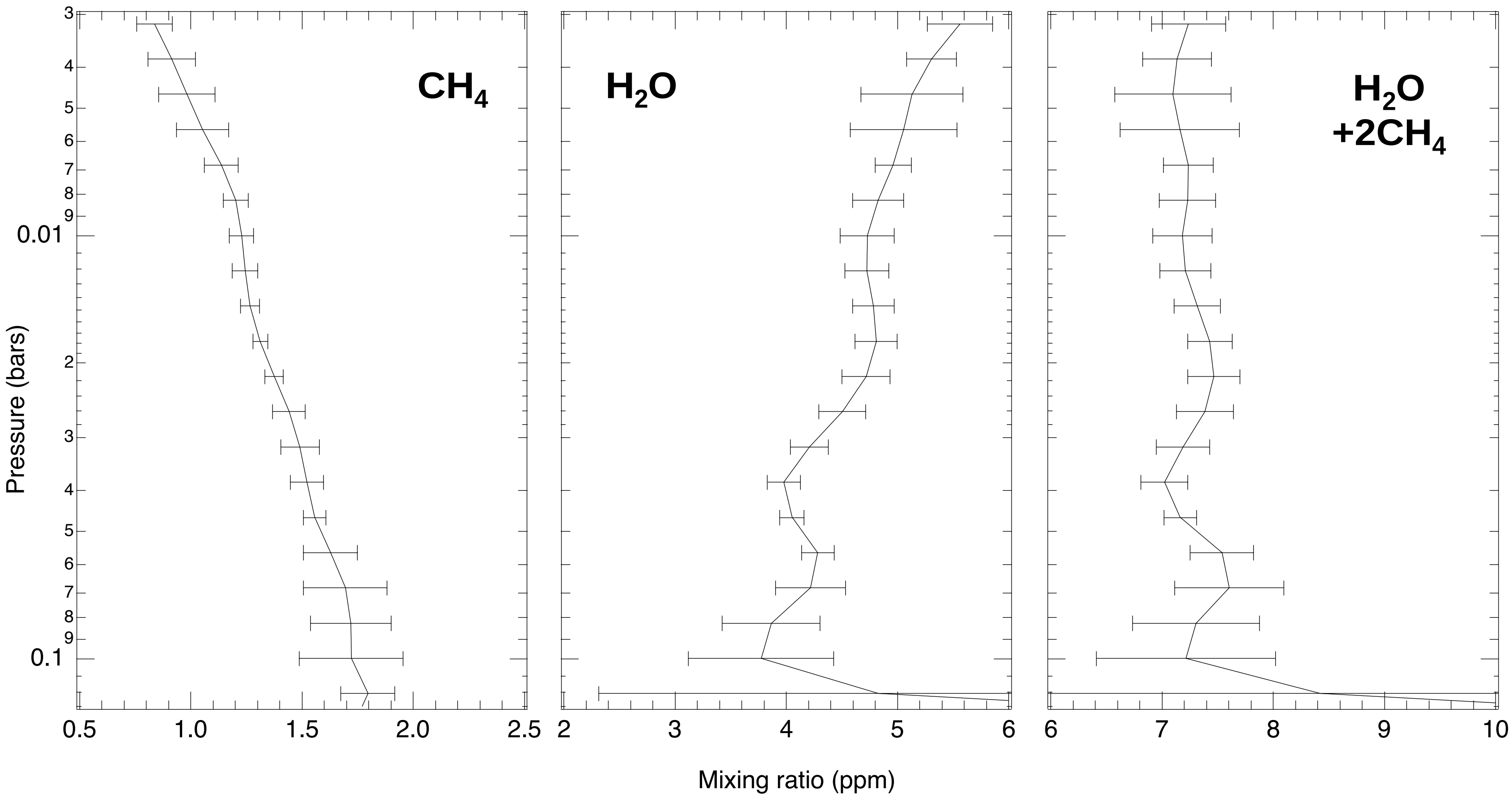

Figure 10: ATMOS/ATLAS-3 measurements of tropical (8°N-20°N) CH_4, H_2O and H_2O+2CH_4. The structure in the H_2O and H_2O+2CH_4 profiles is indicative of the seasonal variation in water injection into the lower stratosphere and its propagation upwards with time. The vertical velocity of tropical upwelling in the lower stratosphere (≈7 km yr^{-1} near 22 km) can be estimated from the wavelength of this structure.

such as the hydrolysis of N_2O_5 to HNO_3, in regulating concentrations of reactive nitrogen (NO_x). Since removal of NO_x serves to increase HO_x and ClO loading in the middle and lower stratosphere, heterogeneous reactions result in a diminished importance of NO_x cycles toward O_3 depletion, contrary to predictions of earlier models that considered only gas phase reactions. ATMOS can provide global measurements of the distributions of NO_x, $ClONO_2$, H_2O, and CH_4, from which the concentrations of HO_x and ClO radicals can be inferred. Measurements of HNO_3, O_3, and aerosol distributions by ATMOS and SAGE III will additionally aid the understanding of the sources and sinks of NO_x .

ATMOS measurements of the halogen-bearing gases demonstrate the overall balance of organic and inorganic chlorine throughout the atmosphere (**Fig. 11**). However, our current understanding is still incomplete. For example, $ClONO_2$ and HCl measured by ATMOS, and ClO measured by ATLAS / MAS (Millimeter-wave Atmospheric Sounder), have revealed inadequacies in the photochemical modeling of reactive chlorine, consistent with a channel forming HCl from the reaction OH+ClO (**Fig. 12**). An ISS deployment of ATMOS and SAGE III will provide measurements of reactive species, their precursors, and aerosols over a range of latitudes, seasons, phases of the QBO, and perhaps after episodic perturbations of the atmosphere such as volcanic eruptions. Analysis of these future observations promises to further our understanding of photochemical processes over a complete range of atmospheric conditions.

Dessler, A. E., D. B. Considine, G. A. Morris, M. R. Schoeberl, J. M. Russell III, A. E. Roche, J. B. Kumer, J. L. Mergenthaler, J. W. Waters, J. C. Gille, and G. K. Yue, Correlated observations of HCl and $ClONO_2$ from UARS and implications for stratospheric chlorine partitioning, *Geophys. Res. Lett.*, *22*, 1721-1724, 1995.

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McElroy, M. B., R. J. Salawitch, and K. Minschwaner, The changing stratosphere, *Planet. Space Sci.*, *40*, 373-401, 1992.

Natarajan, M. and L. B. Callis, Stratospheric photochemical studies with Atmospheric Trace Molecule Spectroscopy (ATMOS) measurements, *J. Geophys. Res.*, *96*, 9361-9370, 1991.

Rinsland, C. P., M. R. Gunson, M. C. Abrams, L. L. Lowes, R. Zander, E. Mahieu, A. Goldman, M. K. W. Ko, D. W. Weisenstein, and N. D. Sze, "Heterogeneous conversion of N_2O_5 to HNO_3 in the post Mt. Pinatubo eruption tropical stratosphere," *J. Geophys. Res.*, *99*, 8213 - 8219, 1994.

Tropospheric Chemistry

In the troposphere, the OH radical is the main oxidizing agent, responsible for removing of non-methane hydrocarbons (NMHCs) and HCFCs before they reach the stratosphere. ATMOS measures many tropospheric species important in production and loss cycles of

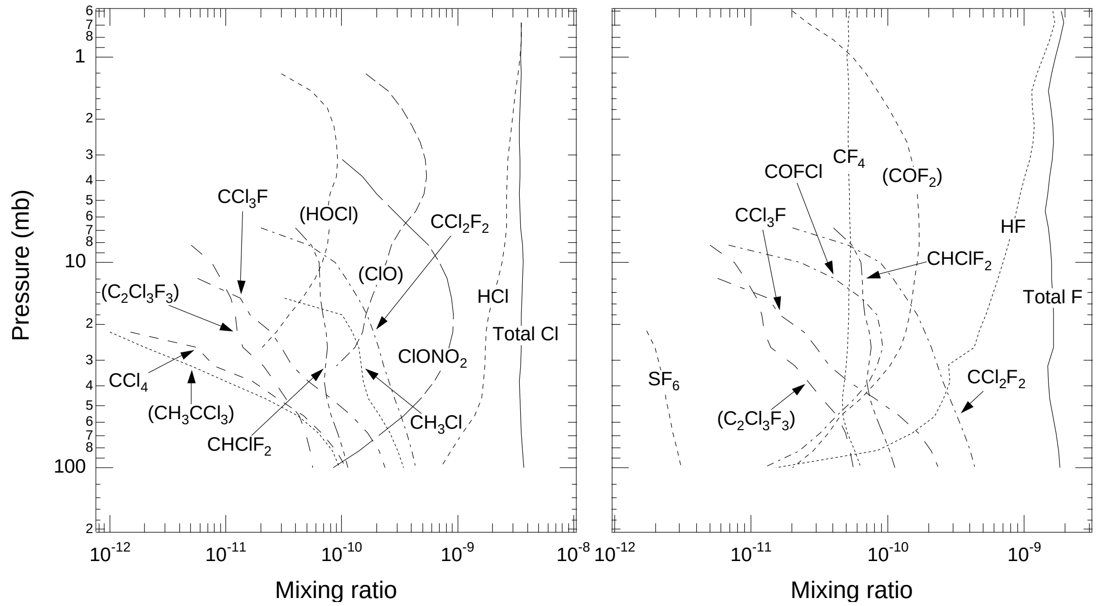


Figure 11: Chlorine and fluorine containing gases measured at northern mid-latitudes from ATMOS/ATLAS-3 and other instruments. The left panel shows the range of source, sink, reservoir and active chlorine species, while the right panel shows those for fluorine species. Species in brackets are profiles from instrument other than ATMOS: ClO from Millimeter Atmospheric Sounder on ATLAS-3 (Courtesy of N. Kämpfer); HOCl from Mark IV FTIR spectrometer balloon measurements near 32 N in 1993 (Courtesy of C. G. Toon), scaled by 3%; CH_3CCl_3 and $C_2Cl_3F_3$ from in situ balloon measurements over France in October 1994 (Courtesy of A. Engel). Mixing ratios of COFCl and COF_2 are 1985 northern mid latitudes values scaled assuming increases of 3% and 2% yr^{-1}, respectively (See Zander et al., *J. Atmos. Chem.*, 15, 171, 1992).

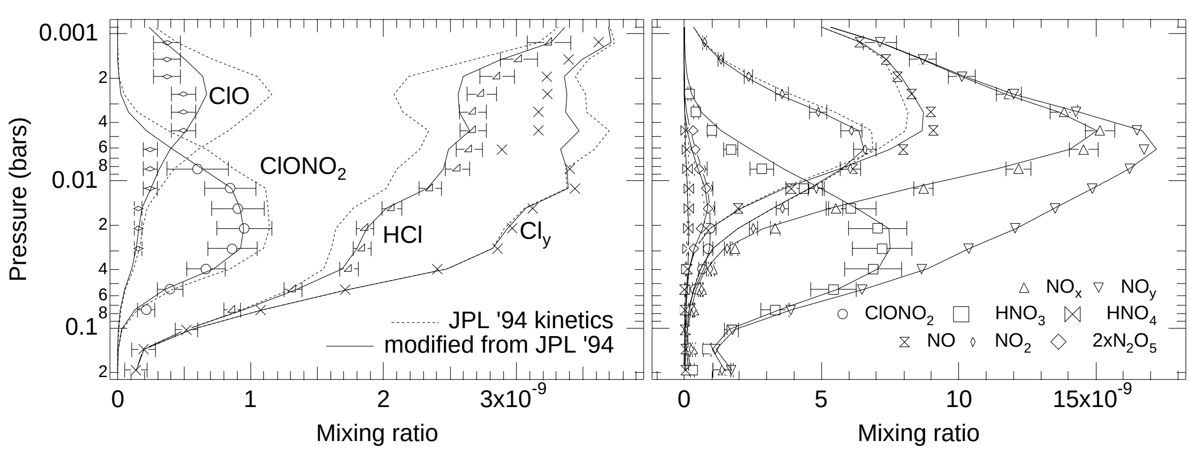


Figure 12: Comparison of measured and modeled inorganic chlorine and odd nitrogen profiles at northern mid-latitudes, early November 1994. The dashed lines represent photochemical model simulations assuming kinetic parameters from DeMore et al. [*Chemical Kinetics and Photochemical Data for Use in Stratospheric Modeling*, JPL Publication 94-26, 1994], while solid lines are model simulations with modifications to the quantum yield of $O(^1D)$ from O_3 photolysis, the HCl branching yield of OH+ClO, and the reaction rates for $Cl+CH_4$ and OH+HCl.

ozone and OH, including O_3, CO, HNO_3, H_2O, and CH_4. ATMOS has measured NMHCs such as C_2H_2 and C_2H_6 to altitudes as low as 5 km. Further examination of the ATMOS spectra is needed to search for signatures of other species which are important at low altitudes, such as H_2CO and HCOOH (which have been measured by infrared solar absorption from balloons). Knowledge of the distribution of H_2CO, which photolyses to OH, is especially crucial for understanding tropospheric distributions of HO_x .

In the recent ATLAS-3 mission, ATMOS was able to acquire a large quantity of high quality spectra well into the troposphere, all the way to the ground. In previous Shuttle missions, these low altitude measurements were rarely obtained due to high aerosol densities and cloud absorption. An ISS deployment will provide a large number of tropospheric observations. Presently, the data reduction algorithms and strategies used by ATMOS have not been optimized for retrieving low altitude data, but proper implementation of the retrieval process for tropospheric studies is achievable (**Fig. 13**).

Rinsland, C. P., R. Zander, C. B. Farmer, R. H. Norton, and J. M. Russell III, "Concentration of ethane (C_2H_6) in the lower stratosphere and the upper troposphere and acetylene (C_2H_2) in the upper troposphere deduced from ATMOS Spacelab 3 spectra," *J. Geophys. Res.*, *92*, 11951-11964, 1987.

Singh, H. B., M. Kanakidou, P. J. Crutzen and D. J. Jacob, High concentrations and photochemical fate of oxygenated hydrocarbons in the global troposphere, *Nature*, *378*, 50-54, 1995.

Polar Processes

The ISS orbit will provide ATMOS with access to latitudes north of ~65˚ N in periods between April and July and south of ~65˚ S between November and January (**Fig. 6**). These summer periods coincide with vortex break-up and chemical recovery of polar air. Past ATMOS missions sampled both the Antarctic and Arctic vortices in their spring period. Analysis of the ATLAS-3 data revealed that denitrification and dehydration associated with sedimentation of stratospheric clouds was confined to air within the Antarctic vortex at all altitudes (**Fig. 14**). Anomalously high concentrations of N_2O_5 were observed for the first time at the vortex edge. The ATLAS-2 observations likewise provided information on the Arctic vortex, still extant in mid-April 1993, following an unusually cold winter that year. Here, evidence of denitrification without dehydration was observed. In both sets of data, the signature of large-scale descent of air within the vortices was observed from the measured profiles of long-lived gases HF, CH_4, and N_2O (**Fig. 15**). The suite of ATMOS species provides diagnostic information on the chemical and dynamical state of the polar atmosphere, and are ideally suited for unraveling the processes of summertime polar vortex recovery.

Fahey, D. W., K. K. Kelly, S. R. Kawa, A. F. Tuck, M. Loewenstein, K. R. Chan, and L. E. Heidt, Observations of denitrification and dehydration in the winter polar stratospheres, *Nature*, *344*, 321-324, 1990.

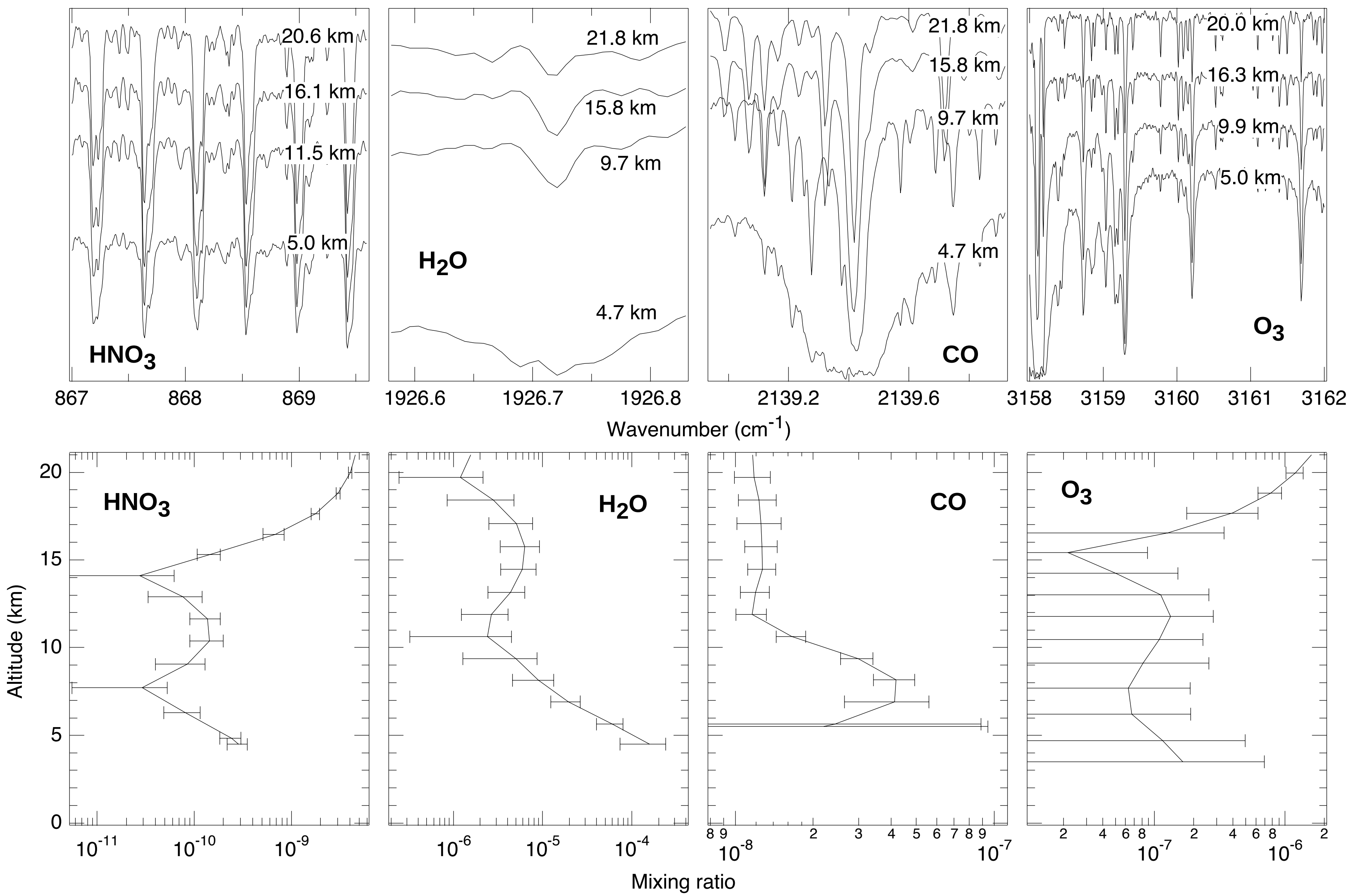
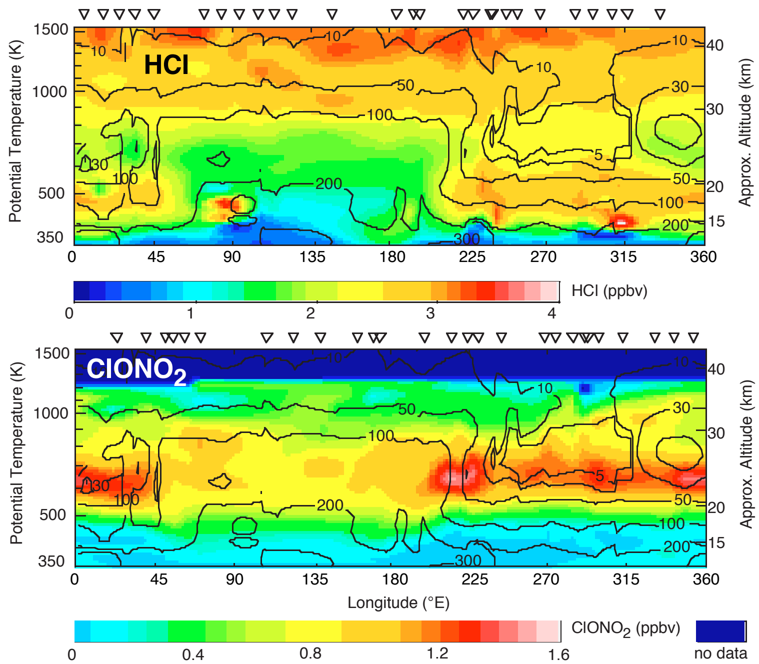


Figure 13: Preliminary lower stratospheric and tropospheric retrievals of HNO_3, CO, H_2O, and O_3. In the future, improved pressure sounding and better spectral window selection are expected to improve low altitude profiles. The upper panels show the corresponding spectral regions used in the retrievals at different tangent heights. Spectra are offset for clarity.



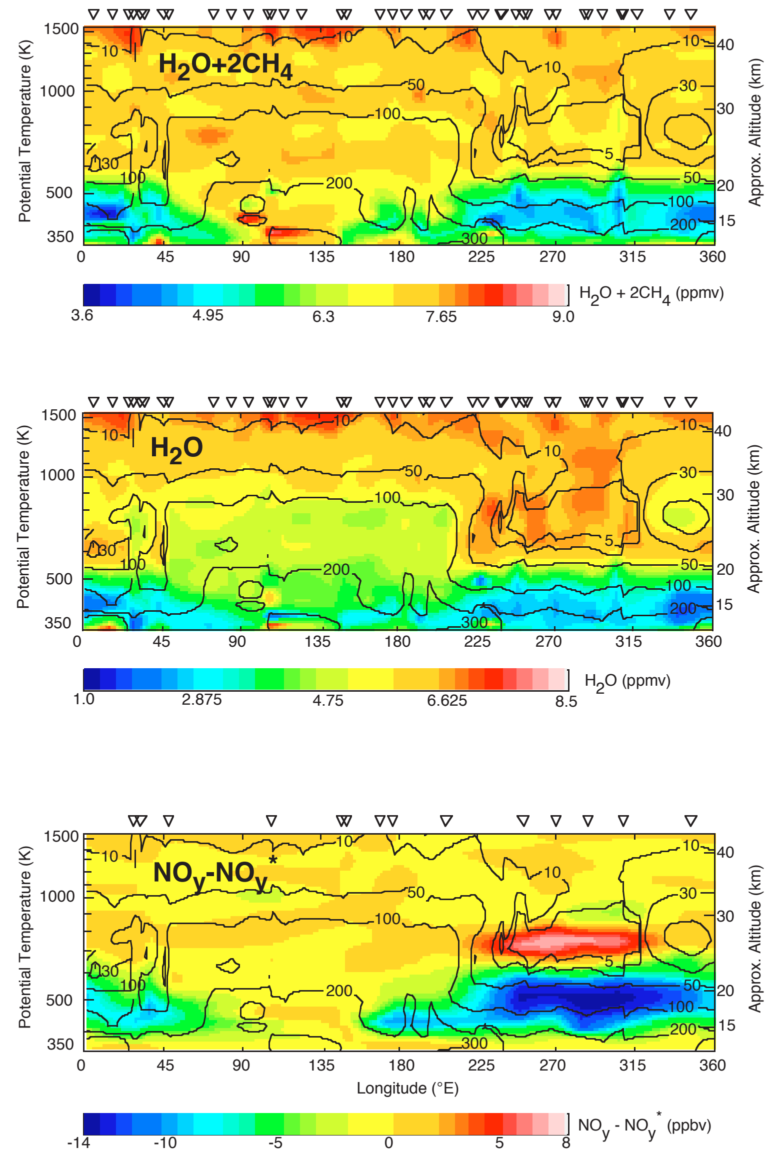


Figure 14: ATMOS measurements of chlorine, hydrogen, and nitrogen species inside and outside the Antarctic polar vortex from November 1994. Panels show [HCl], [$ClONO_2$], [H_2O+$2CH_4$], [H_2O], and [NOy - NO_y*] vs longitude (°E) and potential temperature. Contours of [N_2O] (ppbv) are superimposed: regions of low [N_2O] delineate the polar vortex. [NO_y] = [NO] + [NO_2] + [HNO_3] + 2[N_2O_5] + [HNO_4] + [$ClONO_2$], while [NO_y*] is the predicted abundance of [NO_y] in the absence of denitrification (determined from measurements of NO_y and N_2O outside the vortex). Inverted triangles above each panel mark measurement locations. The observations reveal a region of very high HCl at low altitudes within the vortex, confinement of denitrification and dehydration mainly to the vortex, and a region of enhanced NO_y at high altitude within the vortex that may be due to the production of NO_y from relativistic electrons during polar winter.

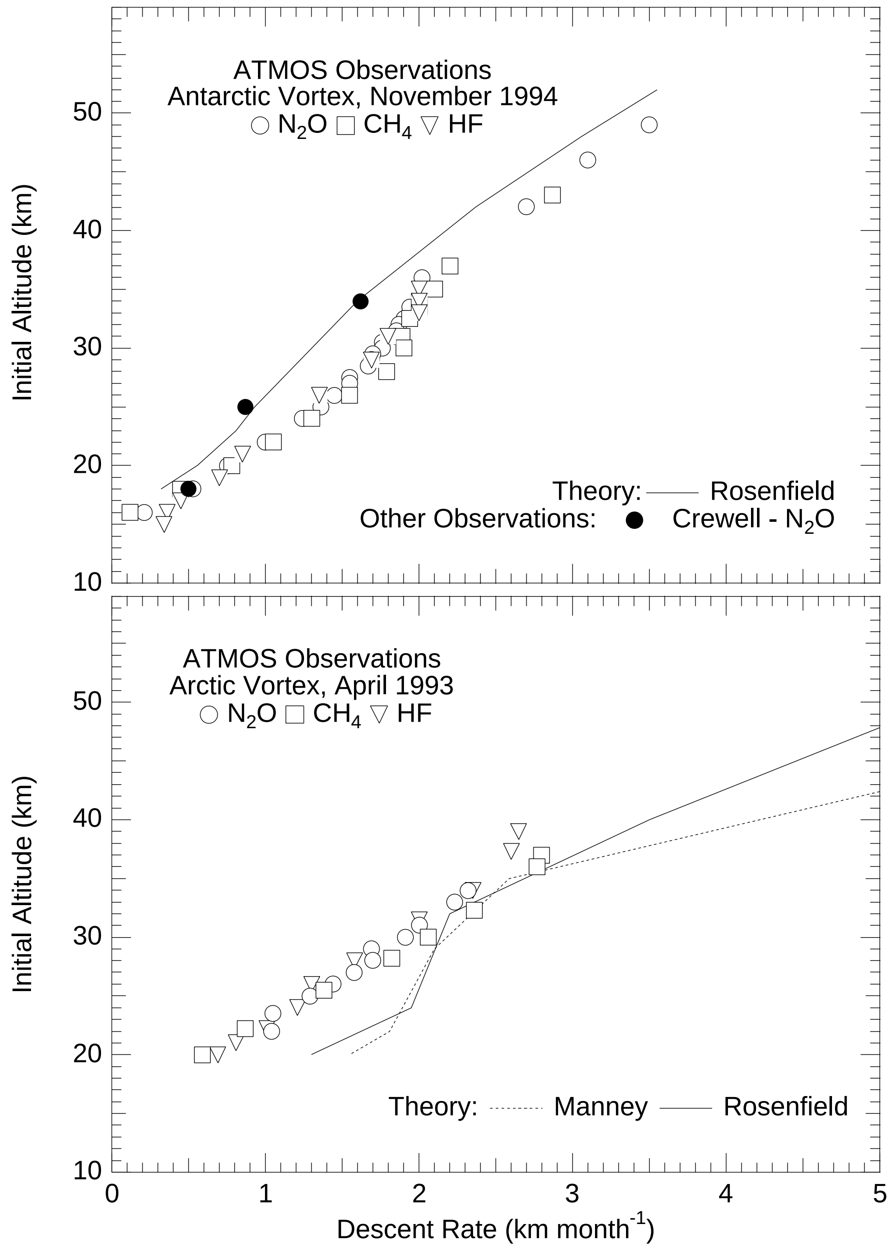


Figure 15: Total inferred descent rates from ATMOS measurements of the Antarctic vortex in November, 1994 (left panel) and the Arctic vortex in April, 1993 (right panel). Rosenfield et al., *J. Geophys. Res.*, 99, 16677-16689, 1994; Crewell et al., *J. Geophys. Res.*, in press, 1995; Bauer et al., *Geophys. Res. Lett.*, 21, 1211-1214, 1994; Traub et al., *J. Geophys. Res.*, 100, 11261-11267, 1995; Schoeberl et al., *J. Geophys. Res.*, 97, 7859-7882, 1992; Manney et al., *J. Atmos. Sci.*, 51, 2973-2994, 1994.

Hoffman, D. J., S. J. Oltmans, B. J. Johnson, J. A. Lathrop, J. M. Harris, and H. Vömel, Recovery of ozone in the lower stratosphere at the South Pole during the spring of 1994, *Geophys. Res. Lett.*, *22*, 2493-2496, 1995.

Schoeberl, M. R., L. R. Lait, P. A. Newman, and J. E. Rosenfield, The structure of the polar vortex, *J. Geophys. Res.*, *97*, 7859-7882, 1992.

Toon, O. B., P. Hamill, R. P. Turco, and J. Pinto, Condensation of HNO_3 and HCl in the winter polar stratospheres, *Geophys. Res. Lett.*, *13*, 1284-1287, 1986.

Radiation and Climate

Climate is directly coupled to the incident flux of solar radiation, the absorption and scattering of this flux, and the trapping of radiation scattered and re-radiated from Earth's surface. One of the key objectives of the ATMOS project remains the survey of the infrared spectral response of the stratosphere, and the evolution of this response with time. The high-resolution, broadband spectra themselves provide a well-calibrated database for assessing the effects of radiatively active trace gases on atmospheric transmission in the thermal IR. Long-term changes in the IR transmission can be evaluated against recorded spectra to ensure the accuracy of the radiation component included in current climate models. The acquisition of ATMOS spectra below the tropopause will provide relative humidity profiles which are of particular relevance toward climate studies. Also the IR detection of various HCFCs in the troposphere is important as these are growing rapidly in concentration and can affect the Earth's radiation balance and climate.

The combination of high spectral resolution IR measurements from ATMOS with co-located UV-VIS measurements from SAGE will represent a unique capability to study the extinction properties of stratospheric sulfate aerosols. These measurements will be especially important in assessing the impact of volcanic aerosols on the radiative budget and consequently on climate. During ATLAS 1, ATMOS spectra were recorded 9 1/2 months after the massive Mt. Pinatubo volcanic eruption and showed strong, broad sulfate aerosol bands throughout the IR below tangent heights of about 30 km. The ATMOS measurements were found to be generally consistent with the shapes, depths, and positions of calculated aerosol spectral bands modeled using the SAGE climatology and SAGE II aerosol observations, thereby confirming that the aerosols were predominantly concentrated H_2SO_4-H_2O droplets. Future ATMOS and SAGE measurements on board ISS will represent an important opportunity to study IR aerosol extinction properties under a wide range of aerosol loadings and atmospheric temperatures.

Rinsland, C. P., G. K. Yue, M. R. Gunson, R. Zander, and M. C. Abrams, "Mid-infrared extinction by sulfate aerosols from the Mt. Pinatubo eruption," *J. Quant. Spectrosc. Radiat. Transfer*, *52*, 241 - 252, 1994.

Yue, G. K., L. R. Poole, P. H. Wang, and E. W. Chiou, Stratospheric aerosol acidity, density, and refractive-index deduced from SAGE-II and NMC temperature data, *J. Geophys. Res.*, *99*, 3727-3738, 1994.

Long-Term Monitoring

ATMOS measurements performed over the lifetime of ISS will provide a long-term record of changes in gases going back to 1985. Although the assessment of changes in atmospheric O_3 itself is provided by the continuous deployment of satellite instruments such as the SAGE series, long-term records of the species that regulate O_3 are needed to further understand the reasons for these changes. Important issues for continued investigation are trends in H_2O and CH_4 which are precursors of HO_x radicals, effects of increased NO_x from exhaust emissions of current aircraft as well as a proposed fleet of stratospheric aircraft, and trends in halogenated gases to ascertain the degree of compliance with legislation to control their abundance.

ATMOS measures a large number of radiatively-active greenhouse gases in the upper troposphere and lower stratosphere, such as O_3, H_2O, CH_4, SF_6, CFC-11, CFC-12, HCFC-22, and N_2O. ATMOS has observed profound increases in the vertical profiles of SF_6 and HCFC-22 (**Fig. 16**). Smaller trends are expected in the profiles of many gases in the middle atmosphere, which may not be fully resolved from ground-based observations. The simultaneous measurement of water and methane profiles, and the ability to make measurements over a full annual cycle, is particularly powerful for studying long-term trends in lower stratospheric/upper tropospheric water vapor.

Accurate monitoring of the concentration profiles of species such as HCl, $ClONO_2$, HF, NO_2, and HNO_3 will improve the interpretation of ground-based column abundances, supporting the efforts of the globally-distributed Network for the Detection of Stratospheric Change (NDSC). ATMOS observations of HCl and HF above 50 km are a measure of total stratospheric chlorine and fluorine (**Fig. 17**) and provide an independent check of the total atmospheric halogen content derived from ground-based *in situ* measurements of source gases. Measurement of changes in the distribution of chlorinated gases in the atmosphere over time will give direct information on the impact of source gas emissions on the chemical composition of the stratosphere. The long-term monitoring of CFC substitutes in the stratosphere and the evaluation of their lifetimes will also ensure the accurate modeling of their effects on climate and ozone.

Cunnold, D. M, P. J. Fraser, R. F. Weiss, R. G. Prinn, P. G. Simmonds, B. R. Miller, F. N. Alyea, and A. J. Crawford, Global trends and annual releases of CCl_3F and CCl_2F_2 estimated from ALE/GAGE and other measurements from July 1978 to June 1991, *J. Geophys. Res.*, *99*, 1107-1126, 1994.

Elkins, J. W., T. M. Thompson, T. H. Swanson, J. H. Butler, B. D. Hall, S. O. Cummings, D. A. Fisher, and A. G. Raffo, Decrease in the growth rates of atmospheric chlorofluorocarbons 11 and 12, *Nature*, *364*, 780-783, 1993.

Gunson, M. R., M. C. Abrams, L. L. L Lowes, E. Mahieu, E., R. Zander, C. P. Rinsland, M. K. W. Ko, N-D. Sze, and D. K. Weisenstein, "Increase in levels of stratospheric chlorine and fluorine loading between 1985 and 1992," *Geophys. Res. Lett.*, *21*, 2223-2226, 1994.

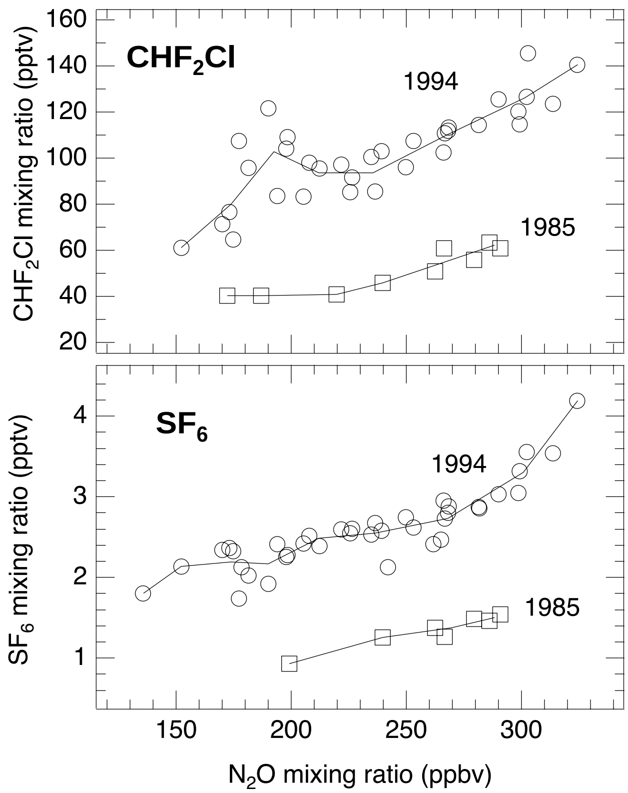


Figure 16: Increase of CHF_2Cl (HCFC-22, upper panel) and SF_6 (lower panel) vs N_2O over northern mid-latitudes. Measurements were by ATMOS from the Spacelab 3 flight of 1985 and the ATLAS-3 flight of 1994. Results indicate an average exponential increase of (7.7±1.2)% yr-1 for CHF_2Cl and (7.1±0.3)% yr-1 for SF_6.

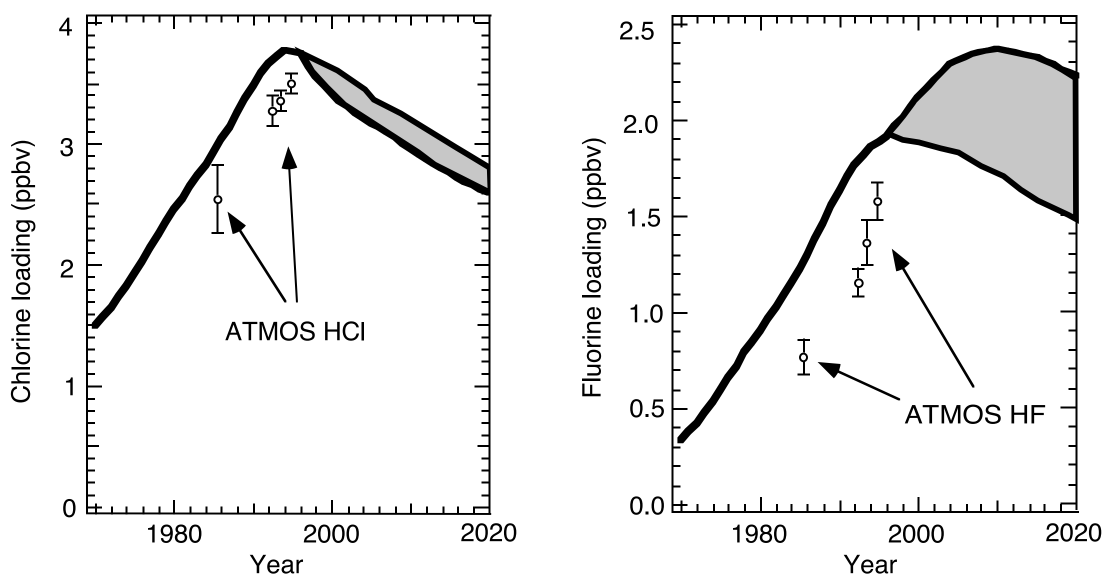


Figure 17: Comparison of measured HCl and HF at 50 km from ATMOS and modeled tropospheric chlorine and fluorine loading. At 50 km, HCl and HF represent essentially all stratospheric chlorine and fluorine, and their mixing ratios at these altitudes provide a sensitive measure of total Cl and F loading in the stratosphere. The modeled curves show the historical increase in Cl and F loading in the troposphere and the predicted future range under various CFC phase-out and replacement scenarios. The differences between the ATMOS measurements and the modeled curves is indicative of the time lag in transport from the troposphere to the upper stratosphere.

Zander, R., M. R. Gunson, C. B. Farmer, C. P. Rinsland, F. W. Irion, and E. Mahieu, The 1985 chlorine and fluorine inventories in the stratosphere based on ATMOS observations at 30° north latitude, *J. Atmos. Chem.*, *15*, 171-186, 1992.

New Species Detection

As the composition of the atmosphere continues to evolve, ATMOS spectra are the ideal means to identify the signatures of new species which may emerge in importance in the next decade, as well as those whose concentrations currently fall below the level of detection, but are expected to continue to increase with time. ATMOS spectra provided the first measurement of atmospheric concentrations of $ClONO_2$, SF_6, COF_2, CH_3Cl, N_2O_5 and HNO_4. As replacement to CFCs increase in abundance in the atmosphere, their detection in the troposphere and lower stratosphere will also be possible through ATMOS spectra. This will be an important part of the assessment of the impact of HCFCs on the stratospheric chlorine and fluorine budget (c. f. **Fig. 18**), and on climate and global change.

The increase in number of ATMOS observations during the ISS deployment will provide high-quality occultation-averaged spectra for improving the detectability of gases limited by signal-to-noise ratio. Averaged spectra in the past have been used successfully to detect changes in SO_2 following the 1992 Mt. Pinatubo eruption. The sum total of data from all Space Shuttle missions was less than 400 occultations; each deployment on ISS should provide on the order of ten thousand occultations per year. Averaging a large number of observations will enable the examination of scientific issues previously limited by retrieval precision (e.g., spatial variations in isotopic enrichments).

Rinsland, C. P., L. R. Brown, and C. B. Farmer, "Infrared spectroscopic detection of sulfur hexafluoride (SF_6) in the lower stratosphere and upper troposphere," *J. Geophys. Res.*, *95*, 5577-5585, 1990.

Toon, G. C., C. B. Farmer, and R. H. Norton, "Detection of stratospheric N_2O_5 by infrared remote sounding," *Nature*, *319*, 570-571, 1986.

Zander, R., C. P. Rinsland, C. B. Farmer, L. R. Brown, and R. H. Norton, "Observation of several chlorine nitrate ($ClONO_2$) bands in stratospheric infrared spectra," *Geophys. Res. Lett.*, *13*, 757-760, 1986.

Correlative Measurements

The detection of trends and global change depends critically on well-validated data records, especially when these are assembled from measurements of disparate instruments. ATMOS measurements constitute one of the best correlative data sources for the comparison against other space-borne, species-specific measurements over time. The ratioing of atmospheric to exoatmospheric spectra, which ATMOS obtains within each occultation period, results in a self-calibrated, fundamental measure of atmospheric transmission as a function of wavelength. Constituent profiles measured by ATMOS do not suffer from long-term

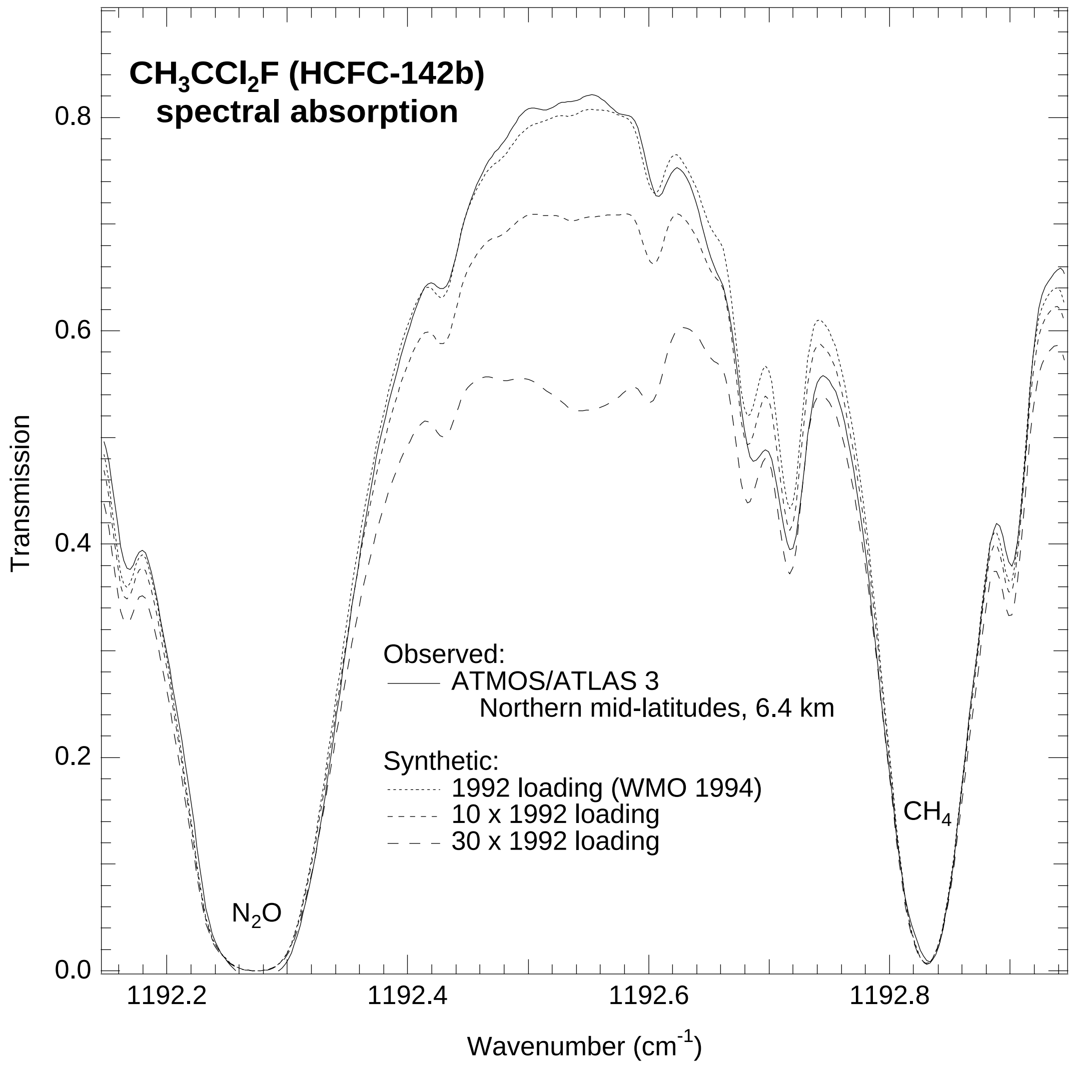


Figure 18. New laboratory cross-sections for the CFC replacement, HCFC-142b (CF_2ClCH_3), will facilitate the measurement of this gas through solar infrared absorption spectra. A November 1994 spectrum from ATMOS/ATLAS-3 at a tangent altitude of 6.4 km is shown, as well as computed spectra assuming HCFC-142b at a 1992 tropospheric mixing ratio of 3.5 pptv (WMO, *Scientific Assessment of Ozone Depletion: 1994,* WMO Report No. 37, 1995), and hypothetically increased mixing ratios of 35 pptv and 105 pptv.

instrument calibration drifts because they do not depend on the detection of absolute radiances. The consistency and inherent completeness of these profiles have been continually demonstrated. In the past, ATMOS correlative measurements have been used in the validation of SAGE II and the instruments aboard the NASA UARS satellite (c.f. **Fig. 19**). ATMOS represent the only correlative measurement source for species such as HCl, HF, $ClONO_2$, CFC-11, CFC-12, and N_2O_5.

The overlap of ATMOS with UARS-generation instruments makes it ideally suited for participation in correlative missions with the NASA EOS Chem-1 satellite, scheduled for deployment in late 2002. On EOS Chem-1, HIRDLS (HIgh-Resolution Dynamics Limb Sounder) will use 21 channels in the IR to profile O_3, H_2O, CH_4, N_2O, NO_2, HNO_3, N_2O_5, CFC-11, CFC-12, $ClONO_2$, and aerosols throughout the stratosphere in emission; an improved MLS will use microwave emission to profile H_2O, O_3, ClO, HCl, OH, HNO_3, NO, N_2O, HF, CO, and SO_2; and TES (Tropospheric Emission Spectrometer), a high-resolution emission FTS operating in both limb and nadir modes, will profile IR active species below 32 km. The significant overlap in species coverage between ATMOS and these various instruments should ensure measurement comparisons of mutual benefit to all platforms.

Atmospheric research satellites planned by other space agencies will also benefit from comparison with ATMOS. These include ILAS on the Japanese ADEOS satellite series (first deployment in 1996), which will measure O_3, HNO_3, NO_2, N_2O, CH_4, and CFC-11 at high latitudes; and those aboard the ESA POEM-ENVISAT series (first launch 1998) such as GOMOS, which will measure O_3, H_2O, NO_2, NO_3, ClO, BrO, OClO, and aerosols; MIPAS, which will measure H_2O, O_3, CH_4, CCl_4, and HNO_3; and SCIAMACHY, which will measure H_2O, O_3, CH_4, N_2O, NO, NO_2, NO_3, ClO, SO_2, BrO, OClO, HCHO, CO, CO_2, and aerosols. Both the ADEOS and POEM-ENVISAT are envisioned as a series of satellites launched at several-year intervals, assuring overlap with an ATMOS deployment on ISS.

Grose, W. and J. Gille, *Upper Atmosphere Research Satellite Validation Workshop III Report: Temperature and Constituents,* NASA Ref. Pub., in preparation, 1995.

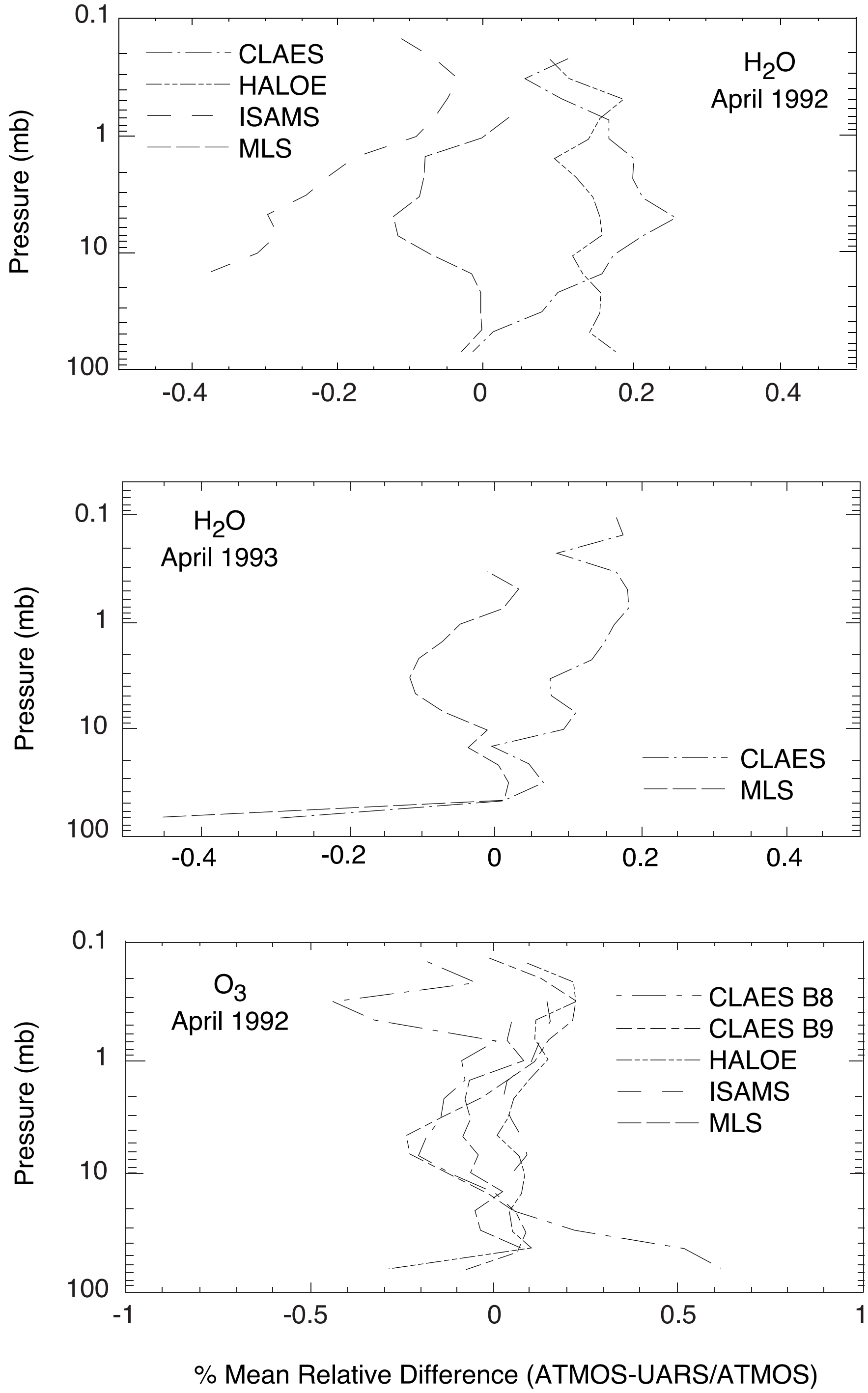


Figure 19: Comparison of spaced-based measurements of O_3 and H_2O from ATMOS and instruments aboard UARS. From Grose, W. and J. Gille, *Upper Atmosphere Research Satellite Validation Workshop III Report: Temperature and Constituents*, NASA Ref. Pub., in preparation, 1995.

Appendix A: Implementation Plan

A detailed Experiment Implementation Plan for the deployment of ATMOS on ISS is being developed. Modifications will be required to the instrument itself, the data reduction procedures, and the data processing facility, but these are expected to be straight-forward.

Deployment Scenario

Optimally, ATMOS will be deployed in a series of 2-year measurement campaigns, with the instrument returning to Earth for ~6 month refurbishments. The first period of deployment would begin in early or mid 2001, and would be scheduled to have at least a 3 month overlap with the EOS Chem-1 satellite. Within each 2 year period, the operational lifetime of ATMOS is expected to be limited by the lifetime of the detector cryocooler system, which, based on vendor specifications (>4000 hrs) and the planned cryocooler operating cycle, is expected to last ~16,000 occultations. This represents 2/3 of all possible occultations over a 2 year cycle. Gaps in observation opportunities are expected due to high beta angle conditions (less than 10%), obstructions in field-of-view by the ISS itself, or possible interruptions in data downlink from the ISS. Limited periods of instrument downtime may be planned to ensure operability for later, more critical times. The refurbishment period will allow replacement of key subsystems to restore the instrument to full functionality.

Instrument Modifications

ATMOS has already demonstrated it can operate in an open space environment and survive the rigors of a Space Shuttle launch. Currently configured to facilitate attachment to the ATLAS pallet of the Shuttle, ATMOS can be readily adapted for use on an ISS Express Pallet. Some physical modifications and refurbishment of the instrument are necessary, mostly to enable the instrument to perform over the 2-year duration of each ISS deployment. Certain subsystems, such as the cryocooler, detector dewar, reference HeNe laser, and power supply will also need upgrading or refurbishment. Radiation shielding for the electronics will be added, and will a passive radiator for providing the necessary heat rejection in the absence of the circulated coolant available on the Space Shuttle. Hardware for data interfaces and on-board data handing may also be required, depending on downlink scenario. In the main, these modifications are expected to be straight-forward and minor, making requalification for space-flight minimal. Preliminary design and planning of these changes has already been initiated.

Resource Requirements

Power requirements for ATMOS, 100 W continuous and 300 W peak, represent a minimal load on the ISS resources. The downlinking of data from the instrument to the ground is presently of some concern. However, the telemetry systems for the ISS have not yet been specified in detail; further information is expected with the release of the ISS Payloads Accommodation Handbook sometime in fiscal year 1996. The ATMOS team is examining data reduction and compression schemes which may provide an affordable relief on the downlink data volume. In past Space Shuttle operations, each ATMOS occultation (comprising ~100 interferograms) contained 500 Mbytes of level 0 data. This data volume would require the ISS data system to handle a load of 16 Mbits per second (Mbps), for the 5 minute duration of each ATMOS occultation, separated by intervals of ~45 minutes. Depending on eventual downlink scenario, some on-board data buffering or processing may be necessary; in the extreme case this would involve converting interferograms (level 0) to spectra (level 1a).

Command and Control

Instrument command and control issues will be addressed to define a process by which instrument control timelines can be automatically generated and modified. During the earlier Space Shuttle missions, continuous human monitoring of the experiment was necessary in order to update instrument commands in response to frequent Shuttle maneuvers and other changes in orientation. This is expected to be much less of a problem on ISS due to more rigidly defined orbital parameters. Nevertheless, instrument operations on ISS must be sustainable without 24 hr human supervision. Any automation of this process in the implementation phase will benefit in lowering manpower costs and increasing efficiencies during the operation phase of the instrument.

Routine Data Processing

During the latest ATLAS-3 mission, a large fraction of the data processing was demonstrated and completed in real time, reflecting the significant improvement in ATMOS data processing since the inception of the project. The first stage of data processing is to convert the raw telemetry data with embedded interferometric values (level 0) to infrared solar power spectra (level 1a). The ratio of the lower tangent height power spectra against an average of exoatmospheric spectra provides atmospheric transmission spectra (level 1b) as the basis for subsequent retrievals. The second stage, which leads to profiles of atmospheric parameters (level 2 data: temperature, pressure and constituent profiles), relies on line-by-line forward model calculations and subsequent inversions based on onion-peeling and nonlinear least-squares spectral fittings. With current computing capabilities, a distributed network of a dozen workstations will be sufficient to handle the data processing required for ISS operations. The continued streamlining and automating the ATMOS data handling process is a priority to ensure the availability of constituent profiles within weeks of data acquisition, both to prevent data backlog and to provide the measurements to the scientific community as rapidly as possible.

Core Science Team

A Core ATMOS Science Team will guide and advise the project on the implementation and operation of ATMOS on ISS. It is envisioned that this will involve the current ATMOS Science Team, which is composed of members from across the US and abroad, to leverage off their expertise and experience; but the Core Team will be augmented in personnel (by a process to be discussed with NASA Headquarters) to maximize the scientific return from an ISS deployment. In the meantime, the current ATMOS Science Team will happily welcome new members and accept general input from the science community, so as to fully exploit the potential of this powerful capability.