

The Atmospheric Trace Molecule Spectroscopy (ATMOS) Experiment:
Experiences from Three Space Shuttle Missions

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Abstract

The Atmospheric Trace Molecule Spectroscopy (ATMOS) project was initiated more than ten years ago, as an investigation of the detailed chemical composition of the middle atmosphere. The required compositional data is obtained through the use of a Fourier transform spectrometer designed to obtain high resolution infrared spectra of the sun during orbital sunrise and sunset on board the Space Shuttle. The instrument has been flown on three Shuttle missions to date: on Spacelab 3 from April 29 to May 7, 1985, on ATLAS-1 from March 24 to April 2, 1992, and on ATLAS-2 from April 8 to April 16, 1993. The technique employed provides the capability to simultaneously measure some thirty atmospheric constituents, including many of the trace species involved in the photochemical processes which control stratospheric ozone levels. The instrument will be described together with the scientific contributions of the ATMOS measurements from these three Shuttle flights.

1. Introduction

For more than a decade, atmospheric scientists have been concerned with the following questions: what is the current state of the Earth's stratosphere, what are the processes involved in establishing this state, and how is it changing? These concerns were brought about originally by the assumed threat to the biosphere as a result of both the depletion of stratospheric ozone caused by anthropogenic emissions of chlorofluorocarbons and the potential for global warming caused by a number of other gases. It has become clear that these are truly "global" problems requiring a global approach, and that studying one particular isolated region can be misleading. For example, extensive studies at midlatitudes failed to predict the recently observed seasonal depletion of ozone in the polar regions and the importance of the role that heterogeneous chemistry is capable of playing in the stratosphere.

The availability of high quality measurements with which to diagnose the dynamical and photochemical processes which are occurring in the stratosphere has proved to be of critical importance; the information acquired in these studies can lead in turn to improved atmospheric models for more

accurate prognoses of future atmospheric evolution. The aim of the Atmospheric Trace Molecule Spectroscopy experiment is to provide simultaneous measurements of more than thirty different atmospheric constituents, under a range of atmospheric conditions and seasons, which can be used to further refine the atmospheric models and improve our understanding of current and future stratospheric conditions.

Over the past twenty-five years at the Jet Propulsion Laboratory, a series of Fourier transform spectrometers have been built and deployed on aircraft and balloons to study the chemical composition of the troposphere and stratosphere. These have been employed primarily in a solar absorption mode; they have been used to measure high resolution infrared solar spectra which contain the absorption signatures of many of the species of interest in stratospheric chemistry. An outgrowth of these efforts was the ATMOS instrument, designed specifically to fly on the Space Shuttle using the Spacelab support systems. On a single mission from low earth orbit, ATMOS can make measurements over broad ranges in latitude, longitude, and altitude, providing global information on the composition of the middle atmosphere.

The ATMOS instrument was built by Honeywell Electro-Optics under contract to NASA and the Jet Propulsion Laboratory (Farmer, 1987; Farmer et al., 1987) using existing 1970s technology; it has been included in the payloads of three Spacelab missions to date. The design philosophy and experiment rationale will be discussed first, and then some of the highlights and scientific results will be discussed in the context of these first three ATMOS flights.

2. Instrument

The ATMOS instrument can be considered as a Connes-type Michelson interferometer built around five major subsystems. It was completed in the early 1980s with "off-the-shelf" hardware conforming to the rather generous weight and power limitations imposed by flying on the Space Shuttle. Twelve years later, both experience and technological advances have suggested that

Table 1- bandpasses of ATMOS optical filters.

Wheel Position	Spacelab 3 (cm ⁻¹)	Typical SNR	ATLAS-1 (cm ⁻¹)	Typical SNR	ATLAS-2 (cm ⁻¹)	Typical SNR
1	600- 1180	300	Same		Same	
2	1100- 2000	150	same		same	
3	1550- 3350	100	same		same	
4	3150- 4800	80	same		same	
5	Double notch [†]	200	600- 2450	150	Same	
6	600- 3500	v. poor	600- 2000	180	1800- 2500	200
7	(not used)		3900-4150	150	600- 1400	250
8	(not used)		600 - 2700	100	2800 - 3100	200

[†] 600-800 and 1800-2500 cm⁻¹

numerous improvements could be made, but in most instances these would be unwarranted as the instrument continues to meet the needs of the ATMOS science team and the community as a whole. It should also be noted that it has been operated with great success to beyond the design life of the subsystems. The minor refurbishments permitted in the time available between successive flights of the instrument have been sufficient to maintain successful operations.

Solar acquisition and tracking during measurement sequences with the ATMOS FTIR is accomplished with a two-axis servo-controlled suntracker, which provides active Sun tracking through 2π steradians with a pointing stability of 0.06 mrad and an accuracy of 0.4 mrad. Sun detection is provided by a silicon array with an acceptance cone angle of 20 degrees. This system relieves the shuttle, a far from perfect platform for precision pointing, of any fine pointing requirements. The reflected beam from the suntracker passes through a ZnSe window onto the instrument foreoptics telescope consisting of two confocal off-axis paraboloids. A selectable field-stop is located at the common focus of these mirrors. For SL-3, these field-stops consisted of 1.0, 2.0, and 4.0 mrad instrument field-of-views, but they were changed to 1, 1.4, 2.0, and 2.8 mrad for the subsequent ATLAS flights. For reference, when the Sun-orbiter tangent point above the limb is 2000 km from the orbiter, the 1 mrad instrument FOV corresponds to a circular area 2 km in diameter. The instrument FOV is selected to optimize signal-to-noise ratios without compromising the spectral resolution (through obliquity effects) or the desired vertical resolution of the retrieved profile, as well as to minimize nonlinearity effects due to detector saturation (Abrams et al., 1993)

The collimated beam from the telescope passes into the interferometer, where cat's-eye reflectors are used in place of the plane mirrors of a classic Michelson

interferometer. These allow the beam to be double-passed through each arm for maximum optical path difference and alignment stability. Solid KBr crystals are used for both beamsplitter and compensator. Both mirrors are moved by a scan-servo mechanism consisting of voice-coil linear motors. Knowledge of the optical path position during a scan is established using reference fringes created by a HcNc laser whose beam traverses the interferometer parallel to the infrared beam. The HcNc reference fringes provide the measurement of the optical path difference (OPD) between the two arms of the interferometer. The HcNc laser is frequency stabilized to a Zecman split single-mode component at 0.633 μ m wavelength. The maximum OPD of 50 cm, corresponding to an unapodized spectral resolution of 0.01 cm⁻¹, is achieved by moving both cat's-eyes in opposite directions over a distance of ± 13 cm, at a mechanical rate of 6.25 cm/sec corresponding to an OPD change rate of 50 cm/sec. The signal from the interferometer is directed through one of eight optical filters and a ZnSe lens onto a HgCdTe detector, which is cooled to 75 K by a split-cycle Stirling mechanical cooler.

The high speed scan-servo system was dictated by a fundamental challenge to the instrument design: at typical shuttle orbital altitudes and inclinations, the sun's rays traverse tangent heights between the surface and 150 km in 1.5 to 2.0 minutes. Consequently, to obtain a high-resolution spectrum every half-scale height of 4 km required a measurement approximately every two seconds. This led to a requirement for very high data sampling rates. At the time of design, a flight qualified 1 MHz, 12-bit analog-to-digital converter was just becoming available, and this provided the crucial component of the ATMOS detector signal chain for facilitating a million point sampling of a double-sided interferogram every two seconds. To obtain an adequate digitization of the full dynamic

range of the **interferometric** signal, two of these ADC'S were incorporated into the signal chain with separate amplification of the signal level. Further signal-to-noise ratio and data rate considerations required that the spectral response of the instrument be limited by optical filters with bandpasses in the 2 to 16pm wavelength range. The ATMOS optical filters used during the SL-3, ATLAS-1, and ATLAS-2 missions are shown in Table 1. Changes were made between missions to optimize the species measurement capability and the signal-to-noise ratio of the observed spectra. This is one example of the benefit of Shuttle-based over free-flying instruments; experience gained during the initial flights can be used in subsequent flights to improve instrument performance and to establish pre- and post-mission calibration procedures to maintain the fidelity of the results.

3. SPACELAB 3 Results

The first ATMOS shuttle mission was as part of the Spacelab 3 payload flown on the Space Shuttle "Challenger", which was launched on April 29th, 1985 (Farmer, 1987; Farmer et al., 1987). A pressure leak in the reference laser housing limited operations of the instrument to a 24 hour period between April 30th, and May 1st, after which the reference laser could not be restarted. Data were obtained through 13 complete atmospheric sunset observations centered around 30°N latitude and six complete sunrise events were obtained around 47°S. During these observations and a number of sun-only viewing periods, a total ~1000 atmospheric spectra and a similar number of sun-only spectra were recorded.

Before analysis, the atmospheric spectra were ratioed against averages of the sun-only spectra to remove spectral features due to the solar atmosphere as well as those arising from residual water vapor and CO₂ inside the interferometer (Norton and Rinsland, 1991). Reference spectral atlases (Farmer and Norton, 1989a,b) have been published of both the sun-only spectra and the atmospheric transmission spectra recorded during the mission. Retrievals of atmospheric geophysical parameters were made using these transmission spectra.

From an orbital altitude of 360 km on SL-3, successive spectra in an occultation were separated in tangent altitude by slightly more than 4 km above 35 km. Below this height, refraction and center-of-brightness effects on the solar disk lead to a gradual reduction down to 1.5 km in spacing of the observations. The vertical resolution in the retrieved profiles of geophysical quantities from these spectra is also limited by the projection of the instrument FOV onto the atmosphere at the tangent

point, as mentioned above.

As a first step to analysis, pressure-temperature information was retrieved (Rinsland et al., 1987; Rinsland et al., 1992) using spectral features of the gases N₂ and CO₂ whose concentrations were well known. The 1-0 vibrational band of the former, with features around 2500 cm⁻¹, provided a reliable means of assigning pressure for spectra between 15 and 35 km tangent heights, while a range of CO₂ features provided an extension of these pressure determinations to heights up to 80 km. High-J lines of the CO₂ v₃ band at 4.3 μm were used to infer temperature. Outside the 10 to 80 km altitude range, recourse was made to climatological data. Over the altitudes of primary interest, the capability of determining pressure-temperature using the minor gases simultaneously with that of abundance of trace gases, provides an internal consistency in the results.

Among the spectral features identified arising from atmospheric constituents were those of a number of gases for which this data set provided the first detection in the middle atmosphere. Included in these were three important reservoir species of the NO_x family (Russell et al., 1988): ClONO₂ (Zander et al., 1986; Zander et al., 1990), N₂O₅ (Toon et al., 1986; Rinsland et al., 1989) and HN₃ (Rinsland et al., 1986). Together with measurements of HN₃, NO₂, and NO, the ATMOS SL-3 data provided a complete inventory of the NO_x family. ClONO₂ links the two catalytic cycles of O₃ destruction involving NO_x and ClO_x families. Although levels of ClO found at mid-latitudes fall below those required for detection in the infrared, ATMOS provided profiles of the final sink atmospheric chlorine (Raper et al., 1987; Zander et al., 1990), HCl, up to heights of 60 km. These measurements, together with the ATMOS measurements of other chlorine-containing gases including all of the most abundant halogen source gases (Zander et al., 1987; Zander et al., 1992.), have provided an estimate of the chlorine loading of the atmosphere for 1985. Indeed, it has been possible from this data to demonstrate that measurements of HCl and HF at 60 km can be used to trace the increases in total -Cl and -F. A summary of the profiles of the main species derived from SL-3 is included in Figure 1. In addition to these species, the quality of the spectra returned permitted the retrieval of the profiles of a large number of isotopes of the minor gases (Gunson et al., 1990; Rinsland et al., 1991) which promise to provide a sensitive means of testing the importance and the rate of atmospheric processes through the resulting fractionation detected in certain isotopomers. The overall set of data has now been

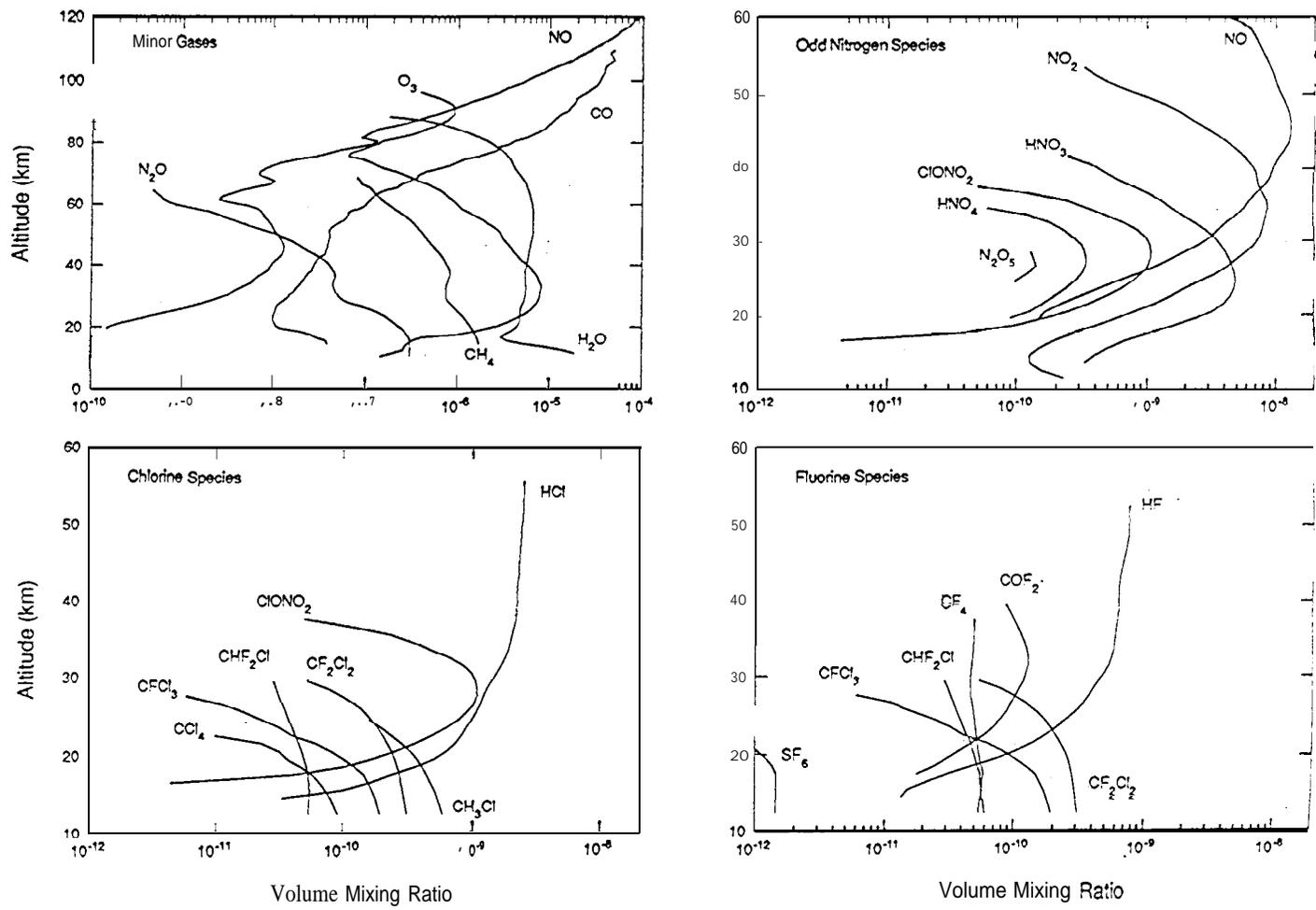


Figure 1 - Summary plot of key profiles measured from ATMOS Spacelab 3 sunset occultation data. These selected profiles represent the four principal groups of species: the minor gases including ozone (Gunson et al., 1990); the NO_x gases (Russell et al., 1988; Rinsland et al., 1989; Zander et al., 1990); Cl-containing gases (Zander et al., 1992); F-containing gases (Zander et al., 1992).

used to evaluate the ability of current photochemical models of the stratosphere to reproduce the observed levels of key gases such as O₃, as well as simpler assumptions such as the steady-state relationships between various species (McElroy and Salawitch, 1989; Naratajan and Callis, 1989; Allen and Delitsky, 1990 and 1991; Pyle and Tuomi, 1990).

4. ATLAS-1 Results

Between March 24th and April 2nd, 1992, ATMOS flew as part of the ATLAS-1 payload on the Space Shuttle "Atlantis". The payload also included a number of other atmospheric remote sensing experiments and solar measurement instruments providing a comprehensive set of measurements for the study of global change. The shuttle was launched at 13:01 GMT into a 57° inclination orbit, and during the flight, ATMOS was successfully operated through some 94 occultation events (Figure 2) covering latitudes from 30°N to 55°S. From the orbital altitude of 300 km, the vertical separation of successive spectra within these observation events was reduced to 3 km as compared to SL-3.

The residual stratospheric aerosol created by the Mt. Pinatubo volcano eruption of July, 1991, caused a diminution of the peak infrared signal detected with all optical filters at tangent heights between 20 and 28 km during the ATLAS-1 flight. The level of attenuation varied greatly between observations with the lowest aerosol levels seen between 20 and 30°S. The optical thickness of this cloud was sufficient at shorter wavelengths to cause the instrument suntracking system, based on a quadrature of photo diodes with responses in the visible, to lose lock on the solar disk through these altitudes. Spectra for these heights show broad spectral features which were not present in the SL-3 spectra, coinciding with extinction caused by H₂SO₄/H₂O droplets (Rinsland et al., 1993a).

Analysis of the ATMOS observations revealed clear evidence of an enhancement in stratospheric HNO₃ beyond those measured in a similar seasonal period by the LIMS instrument on the Nimbus-7 satellite. Prior to the ATMOS measurements, increases in HNO₃ had been seen and explained as evidence of the heterogeneous reaction



Uniquely, ATMOS was also able to measure N₂O₅ simultaneously with HNO₃. In model comparisons, it was evident that observed N₂O₅ profile represented a decrease that could be explained

by inclusion in the model of this reaction (Rinsland et al., 1993b). This is an example of the type of process study to which the ATMOS measurements are well suited, where simultaneous measurement of a number of species can be used to diagnose the magnitude of proposed reaction pathways. Similar studies are underway with the ATMOS measurements to examine the effects of this aerosol in promoting other heterogeneous reactions.

As the ATLAS-1 mission followed some seven years after the first flight of the instrument, significant trends in certain gases were expected from anthropogenic sources. Earlier studies with the ATMOS Spacelab 3 data demonstrated that above 50 km, most of the chlorine released at the surface bound up in halocarbons, is converted into its sink form, HCl. Thus, the measured concentration of HCl at these altitudes can be used as a surrogate for the total chlorine loading of the stratosphere. Confirmation that the source of this increase is a result of the continued release of man-made halocarbons can be found in the measured increases in HF at the same altitudes. With no known significant natural sources for fluorine in the atmosphere, the measured increase in HF can only be as a result of the photochemical breakdown of man-made halocarbons.

An important aspect of the ATLAS-1 mission was its timeliness in furnishing correlative measurements with the Upper Atmospheric Research Satellite suite of instruments (Reber, 1993). In particular, ATMOS has directly comparable measurements to those of the Halogen Occultation Experiment (HALOE) instrument which also operates in the solar occultation mode. Using a 3.5 hour and 2000 km observation matching criteria, there were more than 30 correlative opportunities with HALOE, and using a 3.5 hour and 500 km criteria, there were more than 20 correlative opportunities with the CLAES (Cryogenic Limb Array Etalon Spectrometer) and MLS (Microwave Limb Sounder) limb-viewing UARS experiments. The objective will be for the comparison of the various coincident observations, to maximize the confidence and accuracy in the longer-term global data sets provided by the UARS instruments.

5. ATLAS-2 Preliminary Results

A night launch of the Space Shuttle *Discovery* on April 8, 1993, gave ATMOS on its third flight an opportunity to obtain measurements at orbital sunrise at high northern latitudes (between 60° to 68°N). Although the date of the launch was delayed beyond the critical period when cold temperatures in the lower stratosphere result in the formation of polar stratospheric clouds and the formation of

active chlorine (i.e. Cl₂ and then ClO), the polar vortex was still intact at launch time and was positioned over northern Siberia. These fortuitous circumstances allowed ATMOS to obtain profiles of its suite of gases inside the residual polar vortex as well as outside. Preliminary results indicate that many of the signature conditions identified during the AASE aircraft campaigns were still prevalent in the vortex:

a) low values of the source gases N₂O, CFCs, CH₄ etc. following descent of stratospheric air during the earlier winter period

b) massive conversion of available inorganic chlorine to ClONO₂

This data is currently being analyzed and incorporated into models to test theories of the photochemical formation of, and recovery from the stratospheric polar winter conditions,

6. Future ATMOS Flights

ATMOS is scheduled to fly on the ATLAS-3 shuttle mission in late October, 1994. A flight in this seasonal period will provide ATMOS with an opportunity to obtain measurements at high southern latitudes to complement those made in 1993. The future of this project is uncertain. While it remains a proven, high-quality, high-value research tool in the prime of its scientific life, the changing priorities for the US national space program has left it currently with no manifested payload opportunities.

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