

## ATMOSPHERIC CHEMISTRY EXPERIMENT (ACE): AN OVERVIEW

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### Abstract

*ACE is a Canadian satellite mission that will measure and help to understand the chemical and dynamical processes that control the distribution of ozone in the stratosphere. The ACE instruments are a Fourier transform infrared spectrometer, a UV/visible/near IR spectrograph and a two channel solar imager, all working in solar occultation mode.*

### 1. Introduction

The principal goal of the Atmospheric Chemistry Experiment (ACE) mission is to measure and to understand the chemical and dynamical processes that control the distribution of ozone in the upper troposphere and stratosphere. Anthropogenic changes in atmospheric ozone are increasing the amount of ultraviolet radiation received on the ground and may also affect the climate. A comprehensive set of simultaneous measurements of trace gases, thin clouds, aerosols and temperature will be made by solar occultation from a small satellite in a low earth orbit.

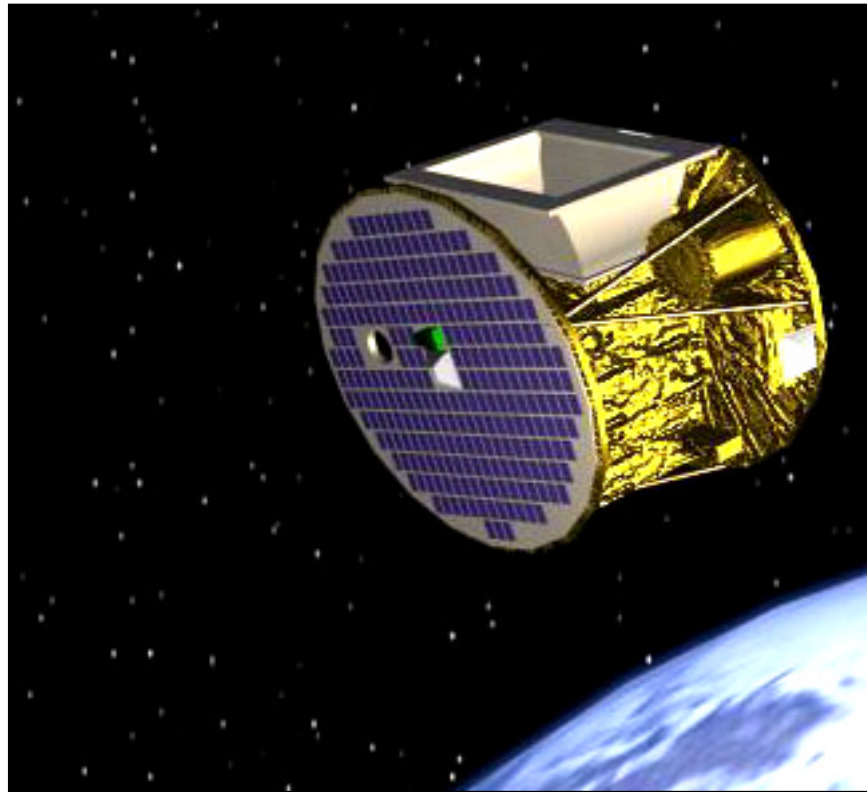
A high resolution ( $0.02\text{ cm}^{-1}$ ) infrared Fourier transform spectrometer (FTS) operating from 2 to 13 microns ( $750\text{-}4100\text{ cm}^{-1}$ ) will measure the vertical distribution of trace gases and temperature. During sunrise and sunset, the FTS measures infrared absorption signals that contain information on different atmospheric layers and thus provides vertical profiles of atmospheric constituents. Aerosols and clouds will be monitored using the extinction of solar radiation at 1.02 and 0.525 microns as measured by two filtered imagers. The vertical resolution will be about 3 - 4 km from the cloud tops up to about 100 km.

A second instrument called MAESTRO (Measurement of Aerosol Extinction in the Stratosphere and Troposphere Retrieved by Occultation) has recently been added to the ACE mission with T. McElroy of the Meteorological Service of Canada (MSC) as the

principal investigator. MAESTRO is a dual optical spectrograph that will cover the 285-1030 nm spectral region. It will have a vertical resolution of about 1 km and will measure primarily ozone, nitrogen dioxide and aerosol/cloud extinction.

A high inclination ( $74^\circ$ ), circular low earth orbit (650 km) will give ACE coverage of tropical, mid-latitude and polar regions. Because reference spectra of the sun will be recorded outside the earth's atmosphere, the ACE instruments will be self-calibrating.

The ACE-FTS and imagers will be built by ABB-Bomem in Quebec City and the satellite bus will be made by Bristol Aerospace in Winnipeg. MAESTRO will be designed and built in a partnership between MSC and the Ottawa-based company EMS Technologies. The satellite will be launched by a NASA using a Pegasus XL rocket in 2002 for a 2-year mission. ACE is the first mission in the Canadian Space Agency's SCISAT-1 program. SCISAT-1 is a small (150 kg) dedicated Canadian science satellite as shown in Figure 1.



*Figure 1.* Artistic view of SCISAT-1 in flight. The circular solar panel and the passive cooler of the FTS (on top) are apparent (from Tom Doherty).

## 2. Science Goals

### 2.1 SCIENCE SUMMARY

#### 2.1.1 *Priority 1*

- a) Measurement of regional polar O<sub>3</sub> budget to determine the extent of O<sub>3</sub> loss. This will require measurements of O<sub>3</sub>, tracers (CH<sub>4</sub> and N<sub>2</sub>O), and meteorological variables (pressure and temperature).
- b) Measurement / inference of details of O<sub>3</sub> budget by detailed species measurements (for O<sub>3</sub>, H<sub>2</sub>O, NO, NO<sub>2</sub>, N<sub>2</sub>O<sub>5</sub>, HNO<sub>3</sub>, HNO<sub>4</sub>, HCl, ClNO<sub>3</sub>, ClO) and modelling.
- c) Measurement of composition, size and density of aerosols and PSCs (Polar Stratospheric Clouds) in the visible, near IR and mid IR.
- d) Comparison of measurements in the Arctic and Antarctic with models to provide insight into the differences, with emphasis on the chlorine budget and denitrification.

#### 2.1.2 *Priority 2*

- a) Mid-latitude O<sub>3</sub> budget.
- b) Measurement of Arctic vortex descent.

#### 2.1.3 *Priority 3*

- a) Study of upper tropospheric chemistry.
- b) Monitoring of CFCs (chlorofluorocarbons), CFC substitutes and greenhouse gases.

### 2.2 DETAILED SCIENCE GOALS

ACE is focussing on one important and serious aspect of the atmospheric ozone problem - the decline of stratospheric ozone at northern mid-latitudes and in the Arctic. Average ozone declines have been measured over much of Canada using ground-based Brewer spectrophotometers [1]. Since 1980 a statistically significant decrease of about 6% has been found by all five long-term Canadian stations (see Figure 2), including Toronto (44°N, 79°W). Ozone sonde measurements show that most of the decline has occurred in the lower stratosphere.

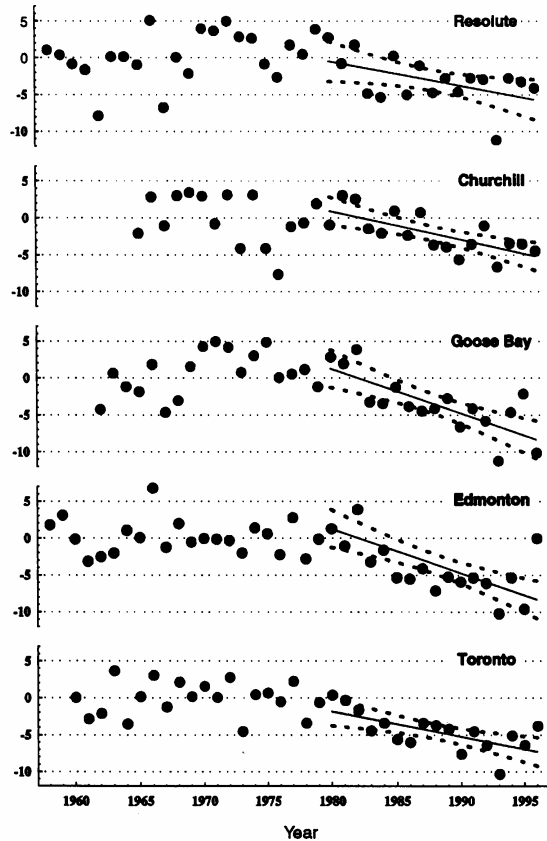


Figure 2. Trends in ozone at Canadian stations [1].

The decrease in Arctic ozone is most severe in the spring as can be seen from the ground and from satellites. The total ozone column obtained from satellite-based TOMS instruments shows this decrease (averaged over the entire polar region) in the month of March (see Figure 3). In March 1997, the ozone column was 21% less than normal and in a small region near the pole the decrease was 40% [2]. Although dynamics plays a major role in redistributing ozone in the Arctic stratosphere, it seems clear that chemical loss of ozone due to heterogeneous chemistry is playing a major role. In fact, several recent publications have indicated that much of the chemical loss in the winter and springtime Arctic is often masked by transport of ozone-rich air from above [3–5]. Specifically, Muller *et al.* [3] estimate that 120-160 DU (Dobson Units) of ozone were chemically destroyed between January and March 1996 overwhelming the dynamical increase leading to a net loss of about 50 DU. This chemical loss is greater than that which was occurring over Antarctica when the ozone hole was first observed in 1985 [6].

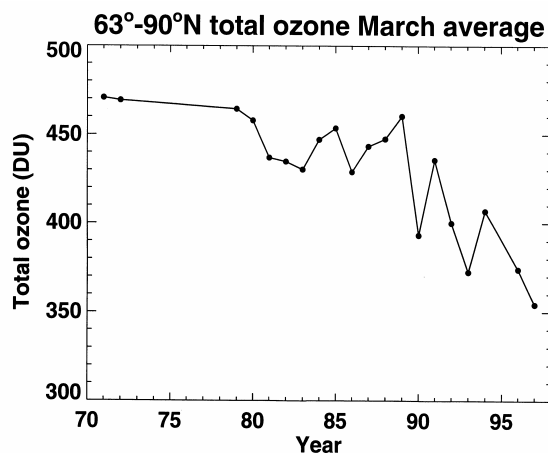


Figure 3. Polar ozone column in March [2].

The major goal of ACE will be to address the question of Arctic ozone loss, and, by means of modelling and measurements (both atmospheric and laboratory) attempt to quantify the contributions from dynamics and chemistry. Since sulphate aerosol and PSCs play a major role in ozone loss in the Arctic, distinguishing between PSCs, aerosols and cirrus and other clouds will be important. There will be an important role for laboratory measurements. The role of modelling, either forward modelling or data assimilation, will also be critical. A variety of atmospheric models ranging from simple box models for the chemistry to atmospheric global circulation models (GCMs) will be used to interpret our data.

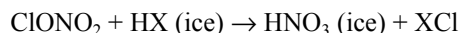
### 2.3 STRATOSPHERIC CHEMISTRY AND ARCTIC OZONE DECLINE

The anthropogenic release of CFCs affects the stratospheric ozone layer through gas phase chemical reactions (such as the Cl/ClO cycle) as well as by heterogeneous chemistry on PSCs and aerosols [1]. CFCs, transported to the stratospheric, are broken down largely by UV light and release “active” chlorine species such as Cl and ClO. These species destroy odd oxygen ( $O + O_3$ ) via various gas phase catalytic cycles. However, the full extent of the destruction is ameliorated by the storage of the active chlorine in the reservoir species HCl and ClONO<sub>2</sub>, neither of which reacts efficiently with O<sub>3</sub> or O. If only the gas phase chemical reactions are considered then the predicted effect on the total ozone column is relatively small. For example, Brasseur [7] predicts with a 2D model (latitude-elevation) that the ozone column would decrease by less than 1% over the ten-year period 1980-1990 in the absence of heterogeneous chemistry.

The discovery of the Antarctic ozone hole in 1985 by Farman *et al.* [6] led to the realisation that heterogeneous reactions were important in determining the ozone budget in polar regions. Later work [8] indicated that heterogeneous reactions on and within sulphate aerosols also play an important role in the stratosphere. Volcanic SO<sub>2</sub> from “random” eruptions penetrating the stratosphere also is important in maintaining the stratospheric sulphate layer budget.

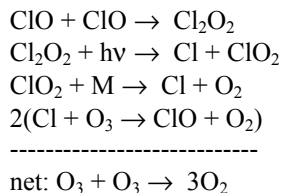
Each winter the vortex forms in the polar regions due to the IR cooling that occurs causing temperatures to drop well below 200 K. The Antarctic vortex is colder than the Arctic vortex. With these cold temperatures H<sub>2</sub>SO<sub>4</sub>, HNO<sub>3</sub> and H<sub>2</sub>O and various mixtures can freeze or exist as supercooled solutions. As the temperature drops below 195 K, large amounts of HNO<sub>3</sub> can dissolve into the sulphate aerosols to form ternary solutions, amounts large enough to deplete almost entirely the gas phase HNO<sub>3</sub> levels. The volume of these aerosol solutions can dramatically increase with this large uptake of gaseous HNO<sub>3</sub> and H<sub>2</sub>O. It is thought that these super-cooled ternary H<sub>2</sub>SO<sub>4</sub>/HNO<sub>3</sub>/H<sub>2</sub>O liquid solutions can exist to within a few degrees of the frost point without forming type I PSC NAT (solid HNO<sub>3</sub>·3H<sub>2</sub>O) and SAT (H<sub>2</sub>SO<sub>4</sub>·4H<sub>2</sub>O). If the temperature falls below the ice frost point (~188 K), type II PSC water ice can form. As temperatures again rise, adsorption of gas phase HNO<sub>3</sub> onto solid SAT can help to regenerate ternary solutions and melt SAT far below the temperature (210-215 K) SAT is believed to melt [9, 10]. Due to the low temperatures needed for solid PSCs to form, it is expected that processing on liquid surfaces is more important in the Arctic than the Antarctic.

Heterogeneous reactions on these condensed phases can activate chlorine and bromine while tying up odd-nitrogen as HNO<sub>3</sub>(a) [11]. The active chlorine and to a lesser extent bromine drive the reactions that form the ozone hole. For example, on the ice crystals, inactive or reservoir forms of the halogen catalysts are freed,

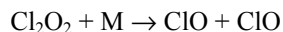


(where X = Cl, Br). Low temperatures drive these processes. As the polar lower stratospheric temperatures drop at the end of the fall season, the aerosol reactions become important. For example, the solubility of HCl is very temperature sensitive and as the temperature drops, it begins to dissolve in the ternary solution / sulphate aerosol. These and similar reactions drive the formation of the ozone hole in the polar late winter (Northern Hemisphere) and austral springtime. These reactions can occur during the night-time and when polar sunrise occurs, species such as Cl<sub>2</sub>, BrCl and ClONO<sub>2</sub> are readily photolysed into more labile species such as Cl. The processed air can persist for several weeks since the reformation of the reservoir species HCl and ClONO<sub>2</sub> is a relatively slow process at polar latitudes. Gravitational sedimentation of the PSCs removes stratospheric HNO<sub>3</sub> and H<sub>2</sub>O (denitrification and dehydration).

One of the main features of ozone loss in polar regions is that it is not rate limited by the low abundance of atomic oxygen. One of main loss mechanisms involves self-reaction of ClO [12]

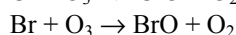
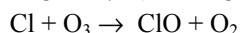
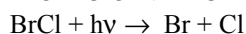
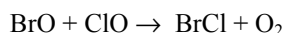
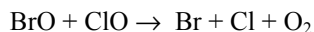


As long as temperatures are less than ~210 K then thermal decomposition of Cl<sub>2</sub>O<sub>2</sub>



which does not lead to ozone loss, does not play a major role.

Another important ozone loss mechanism is due to the synergistic reaction between BrO and ClO [13]:



As before this loss rate is not limited by the abundance of atomic oxygen.

The inclusion of heterogeneous reactions appears to be able to account for the severe ozone loss in the Antarctic spring [1]. There have been problems accounting for the ozone decline in the Arctic [15] although recent modelling work by Lefevre *et al.* [5] using a chemical transport model produces quite good agreement with column ozone measurements. Springtime ozone depletion differs between the Arctic and Antarctic due to differences in seasonal temperature extremes largely caused by atmospheric dynamics related to topography. The northern polar vortex is somewhat larger in extent, less well defined spatially, and more unstable than the southern vortex. More of the northern vortex is exposed to sunlight during the boreal winter, increasing the complexity of both its chemistry and physics. Another effect that may be important is processing that occurs at mesoscales (< 50 km). Most model simulations to date are run at rather low resolution. High-resolution (mesoscale) model runs by Carslaw *et al.* [16] suggest that temperatures sufficiently low for PSCs to form can be induced by mountain waves even when the synoptic temperatures appear to be too high for the formation of PSCs. And as noted above the air, once processed, they will remain processed for several weeks. In this manner large volumes of stratospheric air may undergo processing.

There is no dramatic “Arctic ozone hole” in the spring because downward transport of ozone-rich air masks the strong ozone depletion [3–5]. Additionally, Arctic stratospheric temperatures are generally warmer than those in the Antarctic because the vortex is often in sunlight. The PSCs associated with strong ozone depletion in the springtime form in the stratosphere at temperatures below about 195 K (see above). Such temperatures are common in the Antarctic winter but are rarer in the Arctic. Typical Arctic winter temperatures lie just above 195 K so that there is a strong correlation between springtime Arctic ozone loss and the formation of PSCs. Salawitch has noted the correlation between Arctic ozone column and the minimum temperature measured in March at 50 mbar (about 20 km) (see Figure 4). When this temperature dropped below 195 K as it did in 1994, 1995 and 1997, strong ozone loss was detected. The spring of 1998, however, was relatively warm (198 K) and the ozone loss was much reduced (Figure 4). These observations make a strong case for the importance of heterogeneous PSC chemistry in Arctic ozone declines. In addition to gases, ACE will thus strongly focus on the chemistry and physics of PSC particles. Note that the ozone

declines at mid-latitudes (Figure 2) cannot be attributed to the polar chemistry, although transport of ozone-depleted air might be a contributory factor.

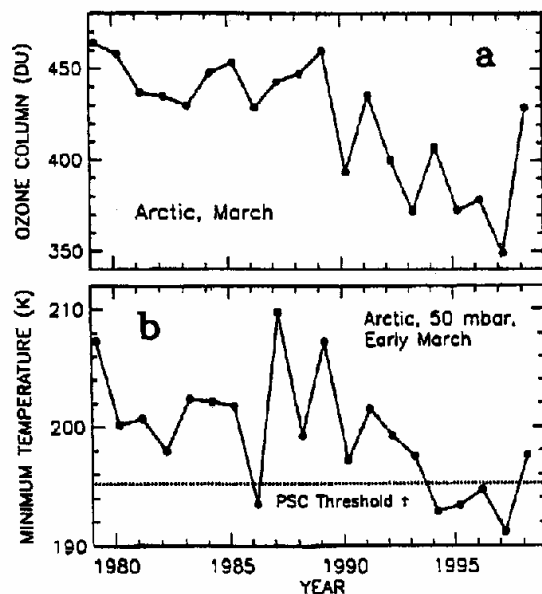


Figure 4. Correlation between minimum Arctic temperature and springtime ozone column (from Salawitch).

#### 2.4 STRATOSPHERIC OZONE LOSS AND CLIMATE CHANGE

Since it was first realised that low temperatures (and high chlorine amounts) drove the polar ozone loss, there has been speculation regarding possible connections between the build-up of  $\text{CO}_2$  and other IR-active gases and polar ozone loss. The increase of these greenhouse gases, while leading to increased IR heating in the troposphere leads to enhanced cooling in the stratosphere and stratosphere temperatures are expected to decrease due to cooling to space from the increased levels of  $\text{CO}_2$ . In fact, this change in the vertical temperature profile has been detected and is taken as strong evidence for a human influence on climate [17]. This increased stratospheric cooling could affect stratospheric dynamics by making the boreal vortex more robust [18] and could lead to an Arctic ozone hole if CFCs continued to increase. The recent work by Shindell *et al.* [19] using a GCM with chemistry suggests that the stratospheric cooling will lead to a prolongation of ozone loss due to cooler temperatures and PSC formation even though chlorine levels decrease.

As noted above, Shindell *et al.* [19] used a Global Climate/Middle Atmospheric Model (GCMAM) with simplified chemistry and constrained ozone transport to investigate the coupling between chemistry and climate. For the period 2010-2019 they found that the increase due to radiative cooling of the stratosphere was 1-2 K poleward of  $70^\circ\text{N}$  while there was an additional cooling of 8-10 K attributable to the increased stability of the Arctic vortex. This dramatic decline in stratospheric temperatures in



their model caused an Arctic ozone hole in the spring with a 2/3 loss of the total ozone column. An Arctic ozone hole is thus predicted to occur in years 2010–2019, in spite of anticipated decreases of CFC concentrations because of the implementation of the Montreal protocol.

The possibility of an Arctic ozone hole is a very disturbing development with strong political and social implications. In contrast to the Antarctic ozone hole, an Arctic ozone hole would affect heavily populated parts of the Northern Hemisphere. Our experimental and theoretical understanding of Arctic chemistry is still in a primitive state. The possibility of the development of an Arctic ozone hole clearly will require more detailed modelling studies combined with a detailed complement of measurements. ACE can contribute to our understanding of physics, chemistry and dynamics of the Arctic polar stratosphere.

### **3. The Orbit**

The selection of an appropriate orbit is critical for the ACE mission to achieve its science goals. Several orbit options were analysed before the baseline mission orbit was chosen during the Phase A period. These orbits included a 57° inclined, 65° inclined, and a 10:30 am sun-synchronous orbit. The baseline orbit was chosen to be circular with an inclination of 65° and an altitude of 650 km.

The sun-synchronous orbit was rejected due to the lack of global coverage. The 57°-inclined orbit was not acceptable due to the lack of polar coverage in the Northern Hemisphere, compared to the baseline orbit. The baseline orbit provided global coverage and coverage of the Arctic region in the spring when ozone depletion is most severe. Ultimately the 65° inclined orbit was shifted to 74° in order to give slightly more polar occultations (at the expense of those in the tropics and mid-latitudes) as well as for easier launch by the Pegasus rocket.

### **4. The FTS Instrument and Imagers**

The main instrument on ACE is a Michelson interferometer with a custom design to meet the requirements of the ACE mission. The interferometer uses two corner cubes rotating on a center flex pivot to produce the optical path difference. A folding mirror inside the interferometer is used to increase the optical path difference. The ACE design is fully compensated for tilt and shear of both moving and stationary optics inside the interferometer. The optical position is measured with a laser diode operating at 1500 nm. The instrument is also equipped with a visible and a near-infrared imager. A pointing mirror, controlled by a suntracker servo-loop, will lock on the sun and track it while the instrument is taking measurements.

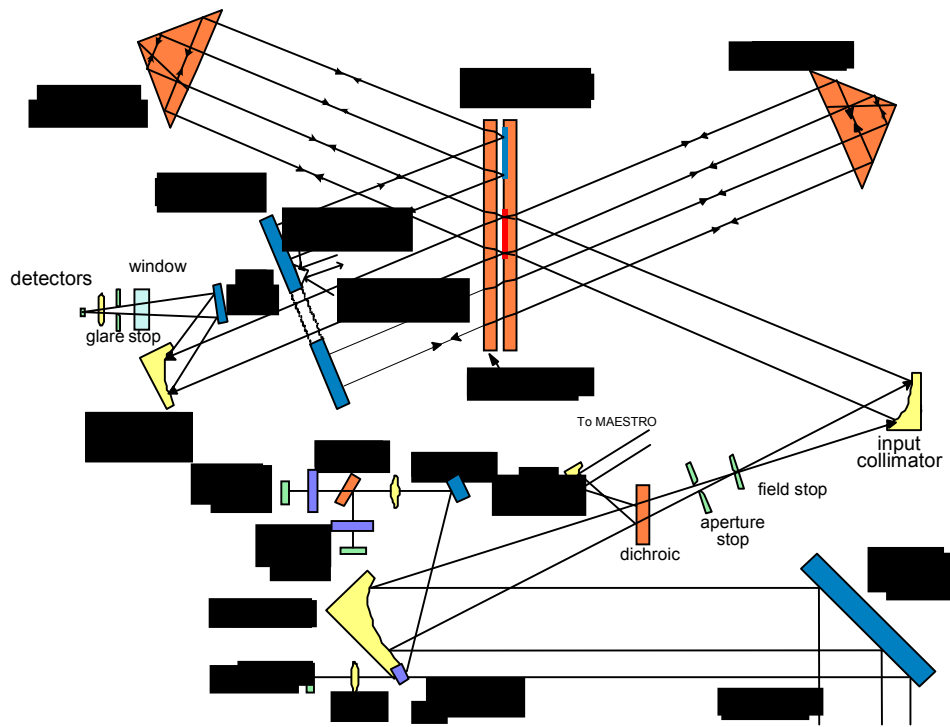


Figure 5. The optical layout of the ACE-FTS and imagers (from ABB-Bomem).

A summary of the main characteristics for the FTS are provided in Table 1.

TABLE 1: Main characteristics of the ACE FTS spectrometer

Spectral range:	750 – 4100 $\text{cm}^{-1}$	Noise equivalent radiance:	< 0.5% of the radiance of a blackbody at 5800 K
Spectral bandwidth:	0.02, 0.04, 0.08, 0.4 $\text{cm}^{-1}$	Detectors:	InSb and HgCdTe
Spectral resolution:	< 0.028, 0.056, 0.11, 0.55 $\text{cm}^{-1}$	Detectors cooling system:	Passive cooling to < 110 K
Sweep duration:	2, 1, 0.5, 0.1 seconds	Field of view:	1.25 mrad
Spectral stability (relative):	$3 \times 10^{-7}$ (rms) for 180 sec.	Mass:	35 kg
Transmittance uncertainty:	< 1% (rms)	Volume:	58440 $\text{cm}^3$
Dynamic range:	0 – 5800 K	Power consumption:	< 40 W

Double-sided interferograms will be Fourier transformed on the ground to obtain the desired atmospheric transmission spectra. The FTS covers the 750-4100  $\text{cm}^{-1}$  range using two detectors (InSb and HgCdTe) in a sandwich configuration. The detectors will

be cooled to less than 110 K (typically 90 K) by a passive radiator pointing towards deep space. The detector/cooler sub-assembly will be built by Ball Aerospace in Colorado.

The satellite will always be pointing towards the sun. This means that during eclipse the FTS will be pointing towards the earth and near-nadir measurements will be possible. Although the signal-to-noise ratio of a single scan will not be high, a rapid series of short scans can be recorded and co-added on the ground.

The visible/near infrared imager has two filtered channels at 0.525 and 1.02  $\mu\text{m}$ , chosen to match two of the wavelengths monitored by the SAGE II satellite instrument [21]. These two wavelengths are useful for the study of clouds and aerosols because they are relatively free of absorption by atmospheric molecules. The detectors in the imagers will be a 256 x 256 active pixel sensors from Fill Factory, a Belgian company. The total field of view of the imagers will be 30 mrad, to be compared to the 9 mrad angular size of the sun. The signal-to-noise ratio of the solar image will be greater than 1000.

## 5. MAESTRO

MAESTRO is a small (about 5 kg) spectrograph that will cover the 285 – 1030 nm region in two overlapping pieces. The use of two spectrographs (280 – 550 nm, 500 – 1030 nm) improves the stray light performance. The spectral resolution is about 1–2 nm and the detectors are linear EG & G Reticon photodiode arrays with 1024 elements. The design is based on a simple concave grating with no moving parts. The entrance slit will be held horizontal to the horizon during sunrise and sunset by controlling the spacecraft roll with a startracker and a momentum wheel on the satellite bus. The FTS, imagers and MAESTRO will all share a single suntracker and will have approximately the same field of view. The vertical resolution of MAESTRO will, however, be about 1 km and will have a signal-to-noise ratio in excess of 1000 as compared to about 200 for the FTS. While the ACE mission will work primarily by solar occultation, MAESTRO will also be able to make some near-nadir solar backscatter measurements like the GOME instrument on the European ERS-2 satellite [22].

## 6. Ground Segment and Data Analysis

The ACE mission is based on the successful (but now retired) ATMOS (Atmospheric Trace Molecule Spectroscopy) instrument which flew four times on the NASA Space Shuttle [23]. ATMOS recorded some remarkable high resolution solar occultation spectra as illustrated by Figure 6. The ACE-FTS instrument has been miniaturized by nearly a factor of 10 in terms of mass, power and volume as compared to ATMOS. Note, however, that ATMOS had a resolution of  $0.01 \text{ cm}^{-1}$  ( $\pm 50 \text{ cm}$  optical path difference) as compared to ACE-FTS ( $0.02 \text{ cm}^{-1}$ ,  $\pm 25 \text{ cm}$  optical path difference).

The ACE mission has been augmented by the two imagers and the MAESTRO spectrograph as compared to ATMOS. By utilizing a small science satellite in a high inclination orbit rather than the shuttle in a low inclination orbit, continuous coverage of

the globe is possible. The ACE orbit improves polar coverage for an occultation instrument.

The imagers will give high signal-to-noise measurements of atmospheric extinction and will lead to SAGE-like aerosol and cloud data products [24]. In addition, the imagers will provide important pointing information low in the atmosphere where refraction and extinction become important. They offer an important diagnostic for the variation of the flux over the distorted solar disk in the lower stratosphere and upper troposphere.

The MAESTRO spectrograph greatly extends the wavelength coverage of the ACE mission. Except for a gap between 1 and 2.4 microns, we will have continuous coverage from 0.28 to 13 microns from the FTS and spectrograph. MAESTRO will measure primarily ozone, nitrogen dioxide and atmospheric extinction, but with a higher signal-to-noise ratio than is possible in the infrared. Moreover, the vertical spatial resolution ( $\sim 1$  km) will be more than 3 times higher than the FTS. This high vertical resolution can be important because there are atmospheric layers (e.g., due to PSCs) that are of order 1 km thick. The wavelength dependence of the atmospheric extinction is useful for determining the aerosol size distribution and particle density. The oxygen A-band at 762 nm (as well as the B- and  $\gamma$ -bands) will be used by MAESTRO to make an independent determination of atmospheric temperature and pressure. It may even prove possible to detect BrO and OCIO in polar regions undergoing perturbed chemistry. The FTS does not have the sensitivity to measure these molecules.

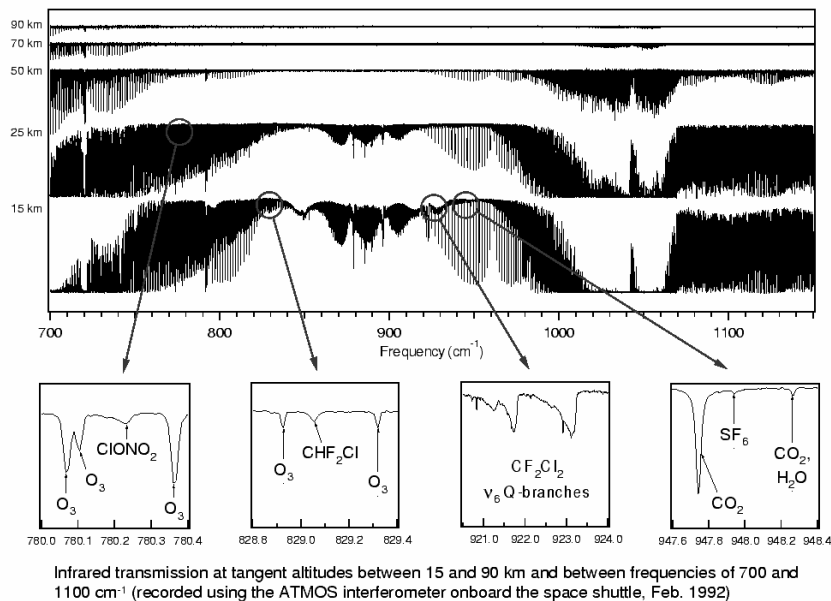


Figure 6. An ATMOS spectrum from C. Rinsland.

The raw ACE data will be sent to ground using at least 2 ground stations. The data volume is about 1 Gbyte per day. These data will be transferred from the Mission Operations Centre operated by Canadian Space Agency in St. Hubert near Montreal to the Science Operations Centre at the University of Waterloo. At Waterloo the data will be archived and transformed into data products for distribution to the science team members. In the case of the FTS, the raw interferograms (level 0) need to be transformed into corrected atmospheric spectra (level 1) by software to be supplied by the instrument contractor, ABB-Bomem.

The atmospheric temperature will be determined as a function of pressure by starting with *a priori* data from a weather forecast model and using a fixed CO<sub>2</sub> volume mixing ratio for the FTS and O<sub>2</sub> A-, B- and  $\gamma$ -bands for MAESTRO. FTS, MAESTRO and imager data will then be converted to height profiles (level 2) of atmospheric species in near real time. In the case of the ACE-FTS, we have inherited the ATMOS code [25] based on an onion-peeling algorithm for the level 1→2 transformation but we will likely use a global fit approach [26]. The MAESTRO algorithms will draw on the previous work by McElroy and co-workers with a variety of UV/visible spectrographs that have been deployed, for example, on the NASA ER-2 aircraft [27]. In the case of the imagers the data products will be SAGE-like extinction profiles [24] as a function of height. Some global data tools (level 3) will be provided for distribution of data over the Web to science team members and the general public.

The ACE mission is completely dependent on the availability of spectroscopic data for the required retrievals of atmospheric molecules and particles. These retrievals are based on the use of a forward model to calculate the radiance incident on the suntracker mirror. Our starting point is the ATMOS linelist [28] which will be up-dated and augmented by the required near UV, visible and infrared cross-sections. This is an enormous task for the members of the ACE science team but is crucial to the success of the mission.

## 7 Acknowledgements

Some of the material in this review paper was taken from the Phase A report prepared by the ACE Science team (see [www.ace.uwaterloo.ca](http://www.ace.uwaterloo.ca) for a list). Many people made important contributions to the Phase A report but J. McConnell (York University) was responsible for much of the material in Section 2. We thank Bomem (M.-A. Soucy) for permission to reproduce Figure 5 and some of the material in Section 4.

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