Atmospheric chemistry experiment (ACE): Analytical chemistry from orbit

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ACE is a Canadian-led satellite mission that is measuring the concentrations of more than 30 atmospheric constituents by absorption spectroscopy using the Sun as a light source. The principal goal of the ACE mission is to make measurements that will improve understanding of the chemical and dynamic processes that control the distribution of ozone in the upper troposphere and stratosphere. The ACE instruments are an infrared Fourier transform spectrometer (FTS), a UV/visible/near IR spectrograph and a two-channel solar imager, all working in solar occultation mode. The high-resolution (0.02 cm^{-1}) FTS is the primary instrument on the satellite, and it covers the mid-infrared spectral region (750–4400 cm^{-1}).

Keywords: Atmospheric science; Fourier transform spectroscopy; Remote sensing; Satellite

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1. Introduction

The Atmospheric Chemistry Experiment (ACE), also known as SCISAT-1, is a small satellite mission for remote sensing of the Earth's atmosphere [1]. The primary goals of the ACE mission are to:

(1) understand the chemical and dynamical processes that control the distribution of ozone in the stratosphere and upper troposphere, particularly in the Arctic;
(2) explore the relationship between atmospheric chemistry and climate change;
(3) study the effects of biomass burning in the free troposphere; and,
(4) measure aerosols and clouds to reduce the uncertainties in their effects on the global energy balance.

We are particularly interested in ozone chemistry because anthropogenic changes in atmospheric ozone have led to an increased amount of ultraviolet radiation reaching the ground and may also affect climate. A comprehensive set of simultaneous measurements of trace gases, thin clouds, aerosols and temperature is being made by solar occultation from a small satellite in low Earth orbit.

A high-resolution (0.02 cm^{-1}) infrared Fourier transform spectrometer (FTS) operating in the range 2–13 microns (750–4400 cm^{-1}) is measuring the vertical distribution of trace gases, particles and temperature. During sunrise and sunset, the FTS measures sequences of atmospheric absorption spectra in the limb viewing geometry with different slant paths and tangent heights; when these spectra are analyzed, the results are inverted into vertical profiles of atmospheric constituents. Aerosols and clouds are monitored using the extinction of solar radiation at 1.02 μm and 0.525 μm as measured by two filtered imagers. The vertical resolution is 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), was 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 covers the 285–1030 nm spectral region. It has a vertical resolution of about 1–2 km and measures primarily ozone, nitrogen dioxide and aerosol/cloud extinction.

A high inclination (74°), circular low Earth orbit (650 km) gives ACE coverage of tropical, mid-latitude and polar regions (Fig. 1). The ACE orbit was selected so that the coverage repeats annually so that Fig. 1 applies every year. Because reference spectra of the Sun (I_0) are recorded

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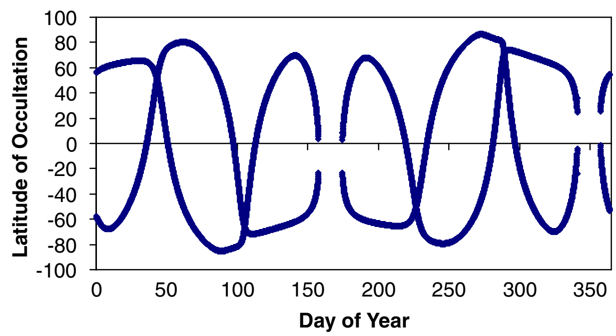

Figure 1. Latitude coverage of the ACE-FTS for one year.

outside the Earth's atmosphere, the ACE instruments are self-calibrating. To be more specific, the observed atmospheric spectra (I) are divided by the appropriate reference solar spectrum and the resulting atmospheric transmission spectra (I/I_0) are analyzed using Beer's Law [2].

The ACE-FTS and imagers were built by ABB-Bomem in Quebec City, Canada, and the satellite bus was made by Bristol Aerospace in Winnipeg, Manitoba, Canada. MAESTRO was designed and built in a partnership between MSC, Ottawa-based company EMS Technologies, and the University of Toronto. The satellite (Fig. 2) was launched by National Aeronautics and Space Administration (NASA) using a Pegasus XL rocket in 12 August 2003 for a nominal 2-year mission. ACE is the first mission in the Canadian Space Agency's small science satellite (SCISAT) program.

The ACE mission is based on the successful (but now retired) ATMOS (Atmospheric Trace Molecule Spectroscopy) instrument that flew four times (1985, 1991, 1992 and 1993) on the NASA Space Shuttle [3]. ATMOS recorded some remarkable high-resolution infrared solar occultation spectra. The ACE-FTS instrument has been miniaturized by nearly a factor of 10 in terms of mass, power and volume compared to ATMOS.

Currently there are two other high-resolution FTSs in orbit:

- the Tropospheric Emission Spectrometer (TES) [4] on NASA's Aura satellite; and,
- the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) [5] on the European Space Agency's ENVISAT satellite.

However, TES and MIPAS monitor the thermal emission of the atmospheric limb (also in nadir mode for TES). Limb-emission spectroscopy offers improved global coverage (day and night) at the expense of signal-to-noise ratio (SNR) and/or spectral resolution as compared to solar occultation.

2. Instruments and analysis

The main instrument on SCISAT-1 is a custom-designed Michelson interferometer (ACE-FTS). The interferometer uses two cube corners rotating on a central flex pivot to produce the optical path difference. An ''end'' mirror inside the interferometer is used to double pass the radiation and increase the optical path difference. The ACE-FTS design is fully compensated for tilt and shear of both moving and stationary optics inside the interferometer. A pointing mirror, controlled by a sun-tracker servo-loop, locks on the center of the Sun and tracks it while the instrument is taking measurements. The FTS has a circular field of view (FOV) of 1.25 mrad, a mass of

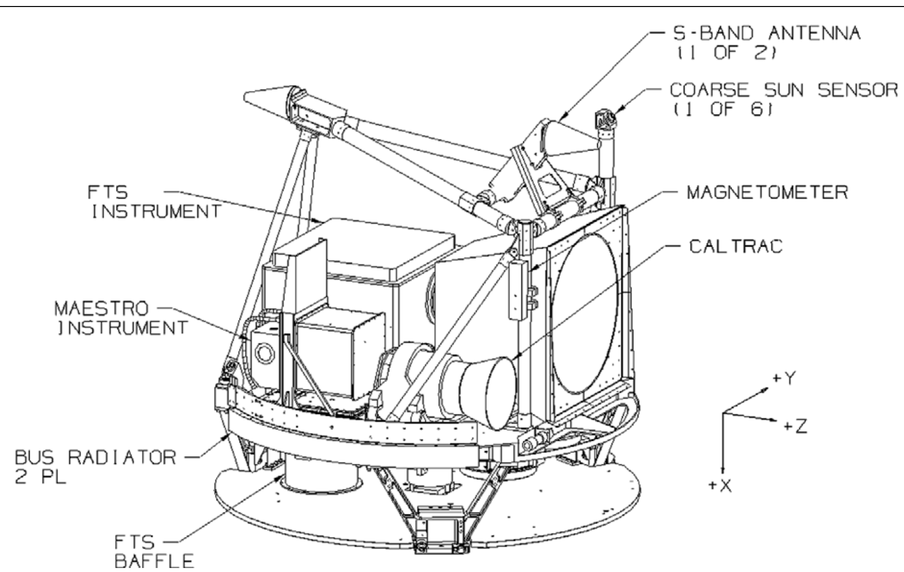

Figure 2. The ACE satellite (SCISAT-1) from Bristol Aerospace. The baseplate is about 1 m in diameter and the total satellite mass is about 250 kg.

about 41 kg, and an average power consumption of 37 W. Double-sided interferograms are Fourier transformed on the ground to obtain the desired atmospheric spectra. The FTS uses two photovoltaic detectors (InSb and HgCdTe), aligned with a dichroic element to have the same field of view. The detectors are cooled to 80–100 K by a passive radiator pointing toward deep space.

The visible/near infrared imager has two filtered channels at 0.525 μm and 1.02 μm, chosen to match two of the wavelengths monitored by the SAGE II satellite instrument [6]. The imagers also provide an important diagnostic for pointing and for detecting the presence of clouds in the FOV. The detectors in the imagers are (effectively) 128×128 active pixel sensors made by Fill Factory of Mechelen, Belgium. The total FOV of the imagers is 30 mrad, to be compared to the 9 mrad angular diameter of the Sun. The SNR of each solar image is greater than 1000, but the main image suffers from overlap by weak secondary images from optical filters that were not tipped far enough off the optical axis.

MAESTRO is a small (about 8 kg) spectrophotometer designed to cover the 285–1030 nm region in two overlapping segments. The use of two spectrographs (280–550 nm, 500–1030 nm) reduces the stray light and permits simultaneous measurements of the two bands with spectral resolution of 1–2 nm, depending on wavelength. The detectors are 1024 linear EG& G Reticon photodiode arrays. The design is based on a simple concave grating with no moving parts. The entrance slit is held horizontal with respect to the horizon during sunrise. The ACE-FTS, imagers and MAESTRO all share a single Sun-tracker and have approximately the same direction of view. The MAESTRO SNR is in excess of 1000. While the ACE mission will work primarily by solar occultation, MAESTRO is also able to make near-nadir solar backscatter measurements, like the GOME instrument on the European ERS-2 satellite [7]. In this review article, we will focus mainly on the ACE-FTS, not MAESTRO and the two imagers.

The ACE-FTS analysis procedure uses the inherent ‘‘self-calibrating’’ advantage of the solar occultation technique. For each occultation, measurements are collected during the time when rays from the Sun pass well above the Earth’s atmosphere (tangent altitudes of 160–225 km), as well as during a brief period when the input mirror is deliberately moved off the Sun to point to deep space. The deep-space measurements are averaged and then subtracted from all ‘‘solar-view’’ measurements for that occultation, in order to correct for self-emission of the instrument. Individual atmospheric measurements (by definition, solar-view measurements with tangent altitudes below 150 km) are divided by an average of the

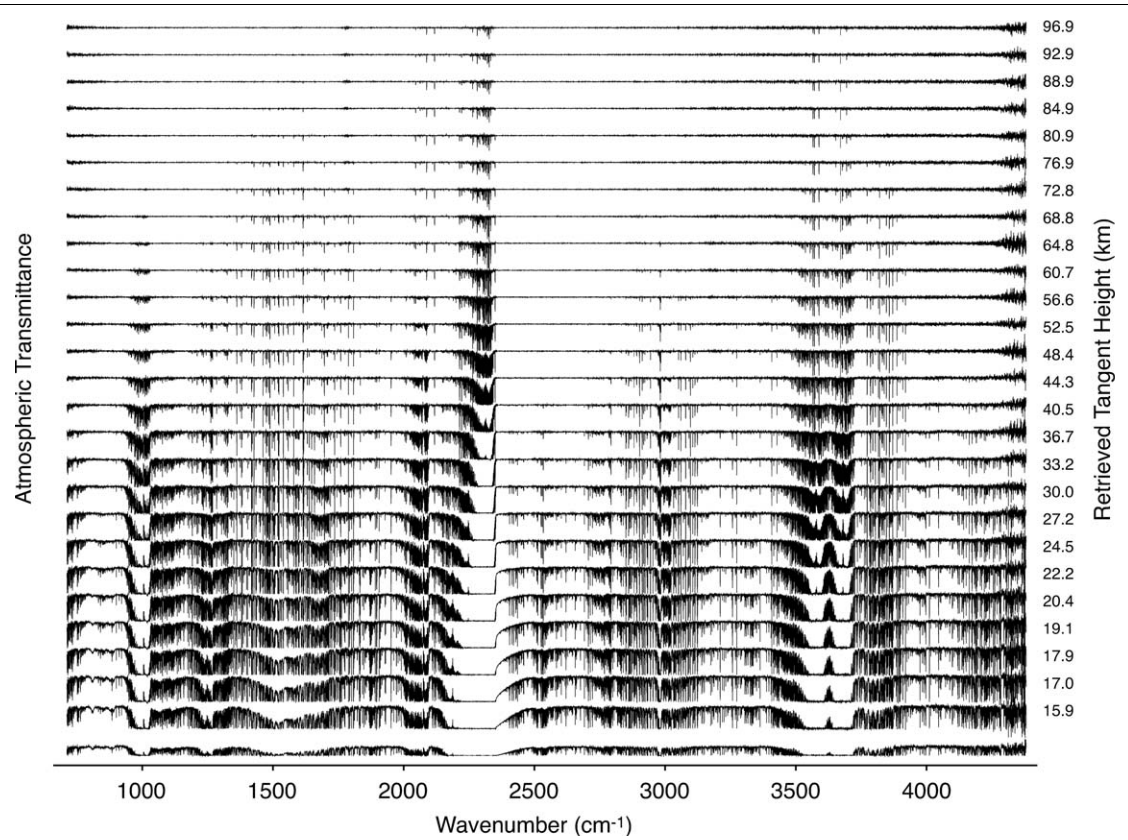

Figure 3. Occultation sequence of atmospheric transmission spectra with the retrieved tangent heights given on the right side.

exoatmospheric spectra for the same occultation, thereby removing both the instrumental response and the solar spectral features. The spectra are recorded every 2 s during sunrise or sunset and a typical sequence of atmospheric transmission spectra is displayed in Fig. 3.

3. ACE-FTS retrievals

The raw ACE data are sent to ground using two Canadian ground stations at St. Hubert (near Montreal, Quebec) and Saskatoon, Saskatchewan, assisted by an American station in Fairbanks, Alaska, USA, and a European station in Kiruna, Sweden. These data are transferred using the internet from the Mission Operations Centre operated by Canadian Space Agency in St. Hubert to the Science Operations Centre at the University of Waterloo. At Waterloo, the data are 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 Fourier transformed into corrected atmospheric spectra (level 1) by software supplied by the instrument contractor, ABB-Bomem. Level 2 data is defined as height profiles of the volume mixing ratios of atmospheric constituents, and is generated by analysis software developed at the University of Waterloo. During level 1 to 2 data processing, the series of spectra measured during an occultation (Fig. 3) is used to infer variations as a function of altitude for the atmospheric quantities of interest.

Prior information on meteorological quantities (i.e. pressure and temperature) is not sufficiently accurate for quantitative analysis of the ACE-FTS measurements, except in the lower troposphere. The first step in the level 1 to 2 data processing is therefore the retrieval of the temperature and pressure profiles for each occultation. Retrievals of temperature from the FTS data assume a fixed CO_2 volume mixing ratio in the altitude range of about 10–70 km. In essence, the relative CO_2 line intensities are used to determine the temperature, and the absolute line intensities give the pressure, so the output is temperature as a function of pressure.

To convert atmospheric pressures into altitudes, we use data from the operational weather analyses [8] of the Canadian Meteorological Centre in a pressure-registration step. A typical example of an atmospheric temperature profile is given in Fig. 4. Note that, below 12 km, we adopt the Canadian weather analyses for our temperature profiles. As the altitude decreases in the troposphere, the ACE temperature retrievals become increasingly difficult because the lines saturate and clouds are often present in the field of view. By contrast, the quality of the temperatures obtained from operational weather analyses improves as the altitude decreases. The 12-km point is a reasonable cross-over altitude to switch from ACE temperatures to CMC temperatures. The error in the ACE temperature retrievals is generally about 1–2 K in the stratosphere and about 1–2% for pressure, although the validation of our retrievals is still in progress.

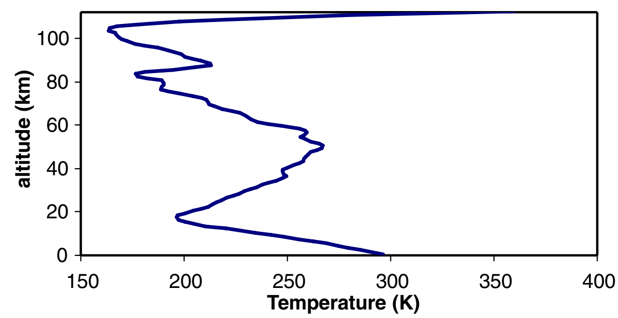

Figure 4. Retrieved temperature profile for the occultation sunset 6500, measured 27 October 2004, latitude 16.9° S, longitude 42.9° E. Note that, below 12 km, temperatures from the CMC weather analyses are used.

The second step in the level 1 to 2 data processing is the retrieval of altitude profiles of the gaseous atmospheric constituents. For the FTS data, the temperature profile is held constant and selected microwindows, typically about 0.5 cm^{-1} wide, are used to retrieve the altitude profiles using a ''global fit'' approach [9]. An example of such retrieval is provided in Fig. 5 for ozone.

The ACE mission completely depends on the availability of spectroscopic data for the required retrievals of atmospheric molecules and temperature. We have adopted the HITRAN 2004 spectroscopic database [10] for the line parameters and cross sections for the processing of ACE-FTS data.

Version 1.0 of the ACE-FTS level 2 data was generated at the University of Waterloo. This data set consists of concentration profiles of 18 molecules: H_2O, O_3, N_2O, CO, CH_4, NO, NO_2, HNO_3, HF, HCl, N_2O_5, $ClONO_2$, CCl_2F_2, CCl_3F, COF_2, CHF_2Cl, HDO, SF_6, plus p and T, interpolated on a 1-km grid for sunrises between February and October 2004. Version 1.0 was intended

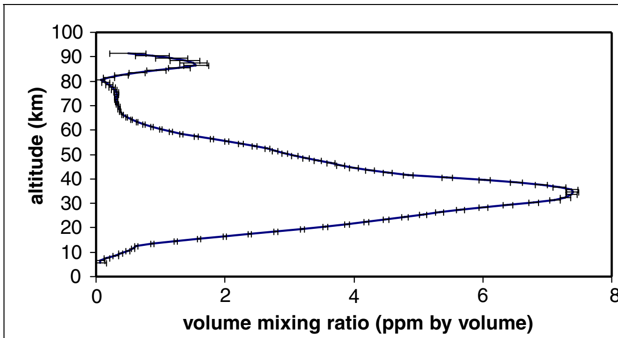

Figure 5. Retrieved ozone profile for the occultation sunset 8430, measured 7 March 2005, latitude 79.8° N, longitude 133° W.

primarily for validation exercises, but was also used for preliminary scientific investigations. A special issue of *Geophysics Research Letters* (http://www.agu.org/journals/ss/ACECHEM1/) was devoted to the first ACE results based on version 1.0 retrievals. The current version is 2.2 for the ACE-FTS processing and now includes the additional molecules HCN, CH_3Cl, CF_4, C_2H_2, C_2H_6, ClO and N_2 (for diagnostic purposes) in the routine processing. The current version of level 2 data for MAESTRO is 1.1, which provides atmospheric profiles of ozone and NO_2. Version 1.0 processing for the imagers is also available and the imager data product is atmospheric extinction at the two imager wavelengths of 525 nm and 1.02 μm.

4. Calibration and validation

The calibration and the validation of the data products provided by a satellite instrument are among the most important tasks in any mission. The results provided by ACE must be compared with those provided by other satellite instruments, ground-based instruments, balloon-borne measurements and the predictions of atmospheric models. This intercomparison is difficult because the various instruments use different techniques and different retrieval algorithms, and the comparison is further complicated by geophysical variability. Comparisons between different instruments require that the measurements be co-incident in space and time. Exact coincidence is not generally possible, so coincidence criteria of typically 500 km in space and 12 hours in time are adopted. One of the most interesting results of the ACE mission will be comparison of O_3 and NO_2 profiles determined by the ACE-FTS and MAESTRO because they are nearly exactly coincident in space and time. The current agreement between ACE and MAESTRO is better than 10% for ozone, but the retrieval algorithms are still being refined.

There are a large number of satellite instruments measuring atmospheric ozone profiles and ACE-FTS (version 1.0) has been compared, for example, with SAGE III, POAM III [11], OSIRIS [12], GOMOS [13] and HALOE [14]. The agreement for ozone is generally within about 10% for version 1.0 and, with further improvements in processing, version 2.2 (''ozone update'') is now agreeing within about 5%. Fig. 6 shows the comparison between ACE version 2.2 ozone update and SAGE III ozone profiles.

The comparisons with the HALOE (HALogen Occultation Experiment) instrument on the UARS satellite are particularly informative. HALOE [14] measures atmospheric profiles of O_3, HF, HCl, H_2O CH_4, NO, NO_2, temperature, and aerosol extinction by solar occultation (like ACE, but mainly using low-resolution infrared filter radiometers, not FTS). The retrieval of a reliable atmospheric temperature profile is crucial because all other retrievals are based on it. The agreement between ACE-FTS and HALOE temperature profiles is very satisfactory and is within 1–2 K [14]. The concentration profiles for the seven molecules measured by HALOE are also generally in good agreement with ACE, except for HCl and HF, for which ACE-FTS volume mixing ratios are 15–20% higher. The origin of this discrepancy is still

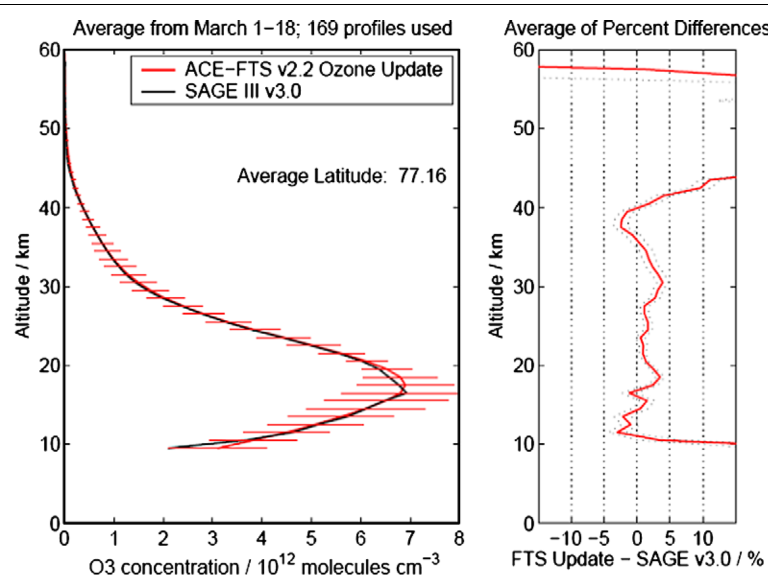

Figure 6. Comparison between ACE-FTS version 2.2 and SAGE III ozone profiles carried out by K. Walker. Notice that, between about 45 km and 60 km, the ozone concentrations are very low and the absolute differences are very small, but the fractional differences are large and off scale in the panel on the right.

under investigation, but the preliminary results are in favor of the higher ACE values.

The Canadian Space Agency has also funded a series of ACE calibration and validation campaigns to Eureka, Nunavut, in the Canadian high Arctic [15]. Some six ground-based instruments were deployed in February–March (2004, 2005 and 2006) to make measurements in coincidence with the ACE satellite. One of the instruments is a copy of the satellite FTS called PARIS-IR [16]. Other ground-based measurements are also being made by various FTSs associated with the Network for the Detection of Stratospheric Change (NDSC, http://www.ndsc.ncep.noaa.gov/), and some preliminary comparisons for HCl and $ClONO_2$ are in satisfactory agreement with ACE observations [17].

5. First results

5.1. Stratospheric chemistry and Arctic ozone declines

The anthropogenic release of chlorofluorocarbons (CFCs) affects the stratospheric ozone layer [18] through gas-phase chemical reactions, such as the Cl/ClO catalytic ozone-destruction cycle:

$$Cl + O_3 \rightarrow ClO + O_2$$
$$ClO + O \rightarrow Cl + O_2,$$

as well as by heterogeneous chemistry on polar stratospheric clouds (PSCs). CFCs are transported to the stratosphere, where they are broken apart by ultraviolet radiation to release ''active'' chlorine species, such as the Cl atom. These species destroy ''odd oxygen'' ($O + O_3$) via various gas-phase catalytic cycles, such as the Cl/ClO cycle. However, the full extent of the destruction is moderated by storage of the active chlorine in the reservoir species HCl and $ClONO_2$, which do not react efficiently with O_3 or O. If only the gas-phase chemical reactions are considered, then the predicted effect on the total ozone column is relatively small.

The discovery of the Antarctic ozone hole in 1985 by Farman et al. [19] led to the realization that heterogeneous reactions were important in determining the ozone budget in polar regions. Each winter, the vortex forms in the polar regions due to the IR cooling that occurs causing temperatures to drop well below 200 K in the stratosphere. These polar vortices are isolated from the rest of the atmosphere (i.e. transport of extravortex air is blocked so that the polar vortices act as isolated chemical reactors). With these cold temperatures, H_2SO_4, HNO_3 and H_2O and various mixtures can freeze or exist as supercooled solutions. As the temperature drops below 195 K, large amounts of HNO_3 can dissolve into the sulfate aerosols to form ternary solutions, amounts large enough to deplete almost entirely the gas-phase HNO_3 levels. The nitric acid can freeze and form solid nitric acid trihydrate (NAT). Type I polar stratospheric clouds (PSCs) are generally thought to be supercooled ternary $H_2SO_4/HNO_3/H_2O$ liquid solutions (Type Ib) or solid NAT ($HNO_3 \cdot 3H_2O$) particles (Type Ia). If the temperature falls below the ice frost point (~188 K), type II PSC water ice can also form in the stratosphere.

Heterogeneous reactions on these condensed phases can activate chlorine and bromine while tying up nitric acid as a solid or in solution. For example, on the ice crystals, inactive or reservoir forms of the halogen catalysts are freed:

$$ClONO_2 + HCl\,(ice) \rightarrow HNO_3\,(ice) + Cl_2(g).$$

Low temperatures drive these processes. These and similar reactions are responsible for the formation of the ozone hole in the polar late winter (Northern Hemisphere) and austral springtime. These reactions can occur during the night, and, when polar sunrise occurs in the spring, species such as Cl_2, BrCl and $ClNO_2$ are readily photolysed into more reactive species, such as Cl. Gravitational sedimentation of the PSCs removes stratospheric HNO_3 and H_2O (called ''denitrification'' and ''dehydration''). The net result of heterogeneous chemistry on PSCs is the conversion of inactive chlorine reservoir species, such as HCl, into active chlorine species, such as ClO, in the polar winter/spring.

One of the main features of ozone loss in polar regions is that it is not limited by the low abundance of atomic oxygen. Atomic oxygen is required for the Cl/ClO catalytic cycle that dominates normal stratospheric chlorine chemistry in the tropics and mid-latitudes. One of main ozone-loss mechanisms that is important only in the polar spring involves self-reaction of ClO:

$$ClO + ClO \rightarrow Cl_2O_2$$
$$Cl_2O_2 + h\nu \rightarrow Cl + ClO_2$$
$$ClO_2 + M \rightarrow Cl + O_2$$
$$2(Cl + O_3 \rightarrow ClO + O_2)$$
$$\text{— — — — — — — —}$$
$$\text{net}: O_3 + O_3 \rightarrow 3O_2$$

The inclusion of heterogeneous reactions appears to be able to account for the severe ozone loss in the Antarctic spring, but the situation is more complicated in the Arctic. Springtime ozone depletion differs between 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, warmer, and more unstable than the southern vortex. More of the northern vortex is exposed to sunlight during the Arctic winter, increasing the complexity of both its chemistry and physics.

There is no dramatic ''Arctic ozone hole'' in the spring, partly because downward transport of ozone-rich air from outside masks the strong ozone depletion [20]. 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.

The ACE mission is unique because it is the only satellite that can monitor all of the main components involved in activated polar chlorine chemistry: O_3, HCl, $ClONO_2$, ClO, HNO_3, H_2O and PSCs. The ACE-FTS measurements of HCl, $ClONO_2$ and ClO were recently used to study chlorine chemistry during the very cold 2005 Arctic winter and spring [21]. In January 2005, PSCs were detected and the concentrations of HCl and $ClONO_2$ reservoir species began to decrease as heterogeneous chemistry proceeded. The minimum value of the HCl volume mixing ratio (vmr) of $\sim$0.2 ppbv was reached around 25 January near about 20 km, and low values persisted until 25 February. After this period of very low levels of HCl, a slow recovery occurred from the end of February until the end of our measurement period at the end of March, when HCl reached values close to those characteristic of unprocessed air masses. For occultations outside the polar vortex, the HCl vmr values at 20.5 km altitude were about 1.5 ppbv while the $ClONO_2$ vmr values were about 0.75 ppbv, with a $ClONO_2$/HCl ratio of about 0.5. Inside the polar vortex, the $ClONO_2$ concentrations covered a wide range, from vmr values as low as 0.1 ppb to values as high as 2.5 ppb during the recovery phase in early March. The ACE-FTS is not sensitive enough to detect the normal background levels of ClO, but can monitor ClO during the periods of enhanced polar chlorine chemistry. For 2005, the maximum vmr value for ClO (about 1 ppbv) was reached near 20 km during the middle of February. These trends in HCl, $ClONO_2$ and ClO were compared with the predictions of a chemical ''box'' model [21] and were found to be generally consistent. Our current understanding of Arctic polar chlorine chemistry and the partitioning of chlorine is therefore satisfactory.

5.2. Long-term atmospheric trends

The potential for recording high-resolution FTS spectra from space was pioneered by the Atmospheric Trace MOlecule Spectroscopy (ATMOS) instrument, which flew successfully during four NASA shuttle flights:

- Spacelab 3 from 29 April –6 May 1985;
- Atmospheric Laboratory for Science and Applications (ATLAS) 1 mission from 24 March–3 April 1992;
- ATLAS 2 mission from 8–16 April 1993; and,
- ATLAS 3 mission from 8–14 November 1994 [3].

The ACE-FTS is now making similar measurements, and, by combining ACE and ATMOS data, changes in the concentrations of atmospheric molecules can be monitored over a 19-year period (1985 to 2004). Long-term trends for the HF, HCl, CCl_2F_2, $CHClF_2$ (HCFC-22), SF_6, CF_4 and H_2O molecules have been derived from vmr profiles measured in the lower stratosphere at mid-latitudes [22–24]. The trend measurements provide evidence of the impact of the emission restrictions imposed by the Montreal Protocol banning the production of CFCs. Decreases in the lower stratospheric mixing ratios of CCl_3F and HCl are measured in 2004 with respect to 1994 [22], providing important confirmation of recent ground-based solar absorption measurements of a decline in HCl. However, the fluorine-containing molecules HF, SF_6, CF_4 and the CFC substitute $CHClF_2$ are still increasing rapidly [22,23].

Water is a crucial molecule in atmospheric chemistry and in climate change. A long-term increase of about 1% per year for water vapor in the stratosphere has been noted by many researchers [25,26]. The combination of ACE and ATMOS results indicates that this steady increase has ceased and the average water-vapor concentration in the stratosphere was about the same in 2004 as it was in 1994 [24].

5.3. Biomass-burning plumes

Biomass burning is a major source of carbon dioxide, methane, nitrogen oxides, and particulate emissions (''pollution''). Agricultural practices result in annual maximum emissions from fires in tropical savannah regions of Africa and South America during August to October. ACE-FTS spectra show elevated levels of the relatively long-lived biomass burning products (CO, C_2H_6, HCN, and C_2H_2) in the upper troposphere and lower stratosphere at 15°S–45°S latitude from 30 September to 3 November 2004 [27]. Mixing ratios up to 260 ppb for CO, 1.47 ppb for HCN, and 1.67 ppb for C_2H_6 are observed in the upper troposphere and their variations are highly correlated, reflecting their similar lifetimes and emission origin. Back trajectory calculations of air-parcel transport and maps of fire distributions for the time period indicated the elevated levels were likely to have originated from regions of tropical fire emissions in South America or Africa with cases identified with elevated emissions reaching close to the lower stratosphere. We have also been able to detect enhanced levels of methanol in these tropical biomass plumes [28]. This is the first time that atmospheric methanol has been detected from orbit.

More recently, we reported on the simultaneous observation of trace gases related to boreal forest fire emissions at 50°N–68°N latitude (29 June–23 July 2004) near Alaska [29]. Mixing ratios for the relatively long-lived biomass burning products (CO, C_2H_6, HCN, CH_3Cl, and CH_4) show plumes with mixing ratios up to 189 ppb for CO, 0.830 ppb for HCN, 1.40 ppb for C_2H_6, 3.00 ppb for CH_3Cl, and 2.09 ppm for CH_4 in the upper troposphere. Elevated tropospheric mixing ratios occur in boreal regions of western Canada and Alaska near those of elevated CO measured from space by the

MOPITT (Measurements of Pollution in the Troposphere) instrument. Back trajectory calculations and maps of fire distributions for the measurement time period indicate the elevated levels measured by both instruments originated primarily from convective transport of the emissions to higher altitudes.

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