Measurements of O₃, NO₂ and Temperature during the 2004 Canadian Arctic ACE Validation Campaign

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[1] The 2004 Canadian Arctic ACE Validation Campaign was conducted to provide correlative data for validating measurements from the Atmospheric Chemistry Experiment (ACE) satellite mission. These measurements were made at Eureka, Nunavut during polar springtime 2004. Six groundbased instruments were operated during the intensive phase of the campaign and ozonesondes and radiosondes were flown. During this time, ACE-FTS and ACE-MAESTRO were performing solar-occultation measurements over the Canadian Arctic. We report the first comparisons between campaign measurements and those from ACE, focusing on O_3 , NO_2 and temperature. Initial mean O_3 profiles from ACE-FTS and ACE-MAESTRO agree to within 20% between 10 and 30 km, and the NO₂ profiles agree to within 40% between 17 and 40 km, which is within the standard deviations. The ACE-FTS temperature profiles agree to better than 2.5 K with the radiosonde temperatures from 10 to 32 km and with the lidar temperatures from 17 to 45 km. Citation: Kerzenmacher, T. E., et al. (2005), Measurements of O₃, NO₂ and Temperature during the 2004 Canadian Arctic ACE Validation Campaign, Geophys. Res. Lett., 32, L16S07, doi:10.1029/2005GL023032.

1. Introduction

[2] The Atmospheric Chemistry Experiment (ACE), also known as SCISAT-1, was launched by the Canadian Space Agency on 12 August 2003 [*Bernath et al.*, 2005]. The goal of ACE is to improve our understanding of the chemical and dynamical processes that control the distribution of O_3 in the upper troposphere and stratosphere. It focuses on the decline of stratospheric O_3 at northern mid-latitudes and in the Arctic. Two instruments make up the ACE payload: a Fourier transform infrared spectrometer, ACE-FTS, and a dual optical spectrophotometer, ACE-MAESTRO (Measurements of Aerosol Extinction in Stratosphere and Troposphere Retrieved by Occultation). These sensors use solar occultation to measure the vertical distribution of atmospheric trace gas species and aerosols.

[3] The 2004 Canadian Arctic ACE Validation Campaign was conducted at Environment Canada's Arctic Stratospheric Ozone Observatory (AStrO) (80.05°N, 86.42°W, 610 m) near Eureka, Nunavut. The intensive phase of the campaign took place from 21 February (polar sunrise at Eureka) to 9 March 2004. Eureka lies directly below the point of maximum stratospheric variability in the Arctic [Harvey and Hitchman, 1996] which makes it an ideal site for stratospheric measurements. The winter polar vortex regularly passes over Eureka and thus measurements both inside and outside the vortex region can usually be made from this single location. During the intensive phase of the campaign, stratospheric temperatures measured by radiosondes remained above 193 K and, according to potential vorticity maps (on the 475 K potential temperature surface) from the European Centre for Medium-Range Weather Forecasts (ECMWF), the measurement site remained outside the polar vortex. In this paper, we focus on comparisons between partial columns and profiles of O₃, NO₂ and temperature retrieved from ACE-FTS, ACE-MAESTRO and the campaign instruments.

2. Instruments and Measurements

[4] During the intensive part of the campaign, six instruments were operated and ozonesondes were launched daily. A Bomem DA8 Fourier transform spectrometer run by the Meteorological Service of Canada (MSC) (MSC-FTS) and a differential absorption lidar (DIAL) are permanently based at Eureka. Four other instruments were deployed at AStrO on a campaign basis. Two of these, PARIS-IR and MAESTRO-G, are ground-based adaptations of the ACE satellite instruments, and the remaining two are UV/visible spectrometers. Nine ACE occultations occurred within 200 km of AStrO during this period (Table 1).

[5] ACE-FTS is a high-resolution (0.02 cm⁻¹) infrared Fourier transform spectrometer, operating from 750– 4400 cm⁻¹, with a two-channel visible/near-IR imager (0.525 and 1.02 μ m) [*Bernath et al.*, 2005; *C. D. Boone et al.*, 2005]. ACE-MAESTRO is a UV/visible/near-IR dual spectrophotometer that measures from 270 to 1040 nm at 1.5–2.5 nm resolution. The ACE-FTS and ACE-MAESTRO measurements are made with a vertical resolution of ~4 km and ~1 km, respectively. The ACE-FTS Version 1.0 data product consists of these results interpolated onto a 1-km grid.

[6] The Portable Atmospheric Research Interferometric Spectrometer (PARIS-IR) is a ground-based adaptation of ACE-FTS with the same spectral range and resolution. The infrared MSC-FTS [*Donovan et al.*, 1997] has been permanently installed at AStrO since 1993. AStrO has been a component of the primary Arctic station of the international Network for the Detection of Stratospheric Change (NDSC)

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OccID	Date and Time, UTC	Latitude	Longitude	Beta Angle ^a	Distance, km
ss2915	26-02 20:00	79.05	-79.4	42.8	179.6
ss2930	27-02 20:26	79.34	-84.6	40.5	87.1
ss2945	28-02 20:52	79.56	-89.4	38.1	79.8
ss2989	02-03 20:32	79.83	-77.1	30.9	183.3
ss3004	03-03 20:58	79.78	-80.5	28.3	119.6
ss3019	04-03 21:24	79.66	-83.7	25.7	68.8
ss3034	05-03 21:50	79.45	-86.8	23.1	67.8
ss3049	06-03 22:16	79.15	-90.0	20.5	123.6
ss3064	07-03 22:42	78.76	-93.2	17.9	199.5

Table 1. Date, Time and Location for the ACE Occultations Within 200 km of AStrO From February-March 2004

^aThe beta angle is the angle between the Earth-Sun line and the orbit of the satellite. The distance between Eureka and the 30 km tangent height subsatellite point is given.

[*Kurylo*, 1991] and this instrument has provided measurements during each spring (and some autumns) for the past ten years. For the Arctic campaign, one suntracker was shared by the two FTSs, so simultaneous measurements were not possible. For O₃ retrievals, the MSC-FTS uses SFIT1 [*Rinsland et al.*, 1982] with microwindows at 782, 2776 and 3040 cm⁻¹ whereas PARIS-IR uses the SFIT2 [*Rinsland et al.*, 1998] multiple microwindow fit with windows at 2775, 2779 and 2781 cm⁻¹. For the MSC-FTS and PARIS-IR NO₂ retrievals, a microwindow at 2915 cm⁻¹ was used with SFIT1 and SFIT2 (retrieving a multiplicative factor for the a priori profile), respectively.

[7] Zenith-sky observations of O_3 and NO_2 were made by three UV/visible spectrometers (MAESTRO-G, the Sun-PhotoSpectrometer and the University of Toronto grating spectrometer) at solar zenith angles (SZAs) up to 96° . The MAESTRO-G is a ground-based version of ACE-MAESTRO with the same spectral range and resolution. The SunPhotoSpectrometer (SPS) [McElroy, 1995] is the forerunner of the MAESTRO instrument. It is a single spectrometer measuring between 295 and 785 nm with a resolution of 1.2-4.0 nm. The SPS also made direct solar observations during the campaign. The University of Toronto triple-grating spectrometer [Bassford et al., 2001, 2005] was operated with a spectral range of 325 to 680 nm and a resolution of approximately 1.5 nm. The grating spectrometer measurements were analyzed using the Differential Optical Absorption Spectroscopy (DOAS) technique. Herein, the grating spectrometer will be referred to as the DOAS.

[8] O_3 and temperature profiles were measured with the DIAL. It is a 5-channel lidar which uses a XeCl laser with hydrogen Raman shifter to provide outputs at 308 and 353 nm [*Bird et al.*, 1996]. A 1-m Newtonian telescope collects the elastic backscattered radiation at these wavelengths as well as the Raman scattering from atmospheric nitrogen and water vapor. The resolution of the profiles is 1.3 km below 30 km, quadratically increasing to 5 km at 50 km and 5 km thereafter. At the Eureka weather station, weekly O_3 measurements are made by MSC using ozonesondes [*Davies et al.*, 2000]. Ozonesondes were launched daily from 24 February to 8 March 2004. Radiosondes were launched routinely twice a day. The profiles have a vertical resolution of less than 50 m.

3. Results and Discussion

[9] This paper presents first results from the campaign and focuses on O_3 , NO_2 and temperature. For the MSC-FTS and PARIS-IR, daily mean total columns were calculated for each species. Weighted averages were calculated for PARIS-IR, where the measurement error determined from the averaging kernel and the fitting error were used as the weighting factors for O_3 and NO_2 , respectively. For PARIS-IR, the uncertainties reported are one standard deviation for the daily average value.

[10] The error estimates for the MSC-FTS were obtained by using the procedure of *Murphy et al.* [2001] which accounts for effects due to the instrument, choice of microwindows, line parameters, temperature profiles, a priori volume mixing ratio profiles, spectral signal-to-noise ratio, and sky condition variability.

[11] The three UV/visible spectrometers record twilight spectra of sunlight scattered from the zenith-sky, from which O_3 and NO_2 slant column densities are retrieved relative to a reference spectrum. Then vertical column densities are retrieved using airmass factors (AMFs) calculated with a radiative transfer model [*McLinden et al.*, 2002]. The AMF at 90° SZA for NO₂ was 17.137 and for O_3 was 16.924. The error estimates for the DOAS were derived by *Bassford et al.* [2005] for mid-latitude conditions and modified for Arctic conditions to yield 8% for O_3 and 15% for NO₂. The MAESTRO-G and SPS O_3 columns reported here have estimated errors of 5%.

3.1. O₃

[12] Figure 1 shows the partial column densities for ACE-FTS, ACE-MAESTRO, the DIAL and the ozonesondes, obtained by integrating the number density profiles from 10 to 30 km (altitudes for which data exist for all four instruments). Retrieved mixing ratios from ACE-FTS and



Figure 1. O_3 partial columns (10–30 km) measured by ACE-FTS, ACE-MAESTRO, the DIAL and the ozone-sondes.

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(a)



Figure 2. O_3 total columns measured by the three UV/ visible spectrometers and the two ground-based FTSs, and the derived ACE-FTS total column. The line shows the ACE-FTS variation.

ACE-MAESTRO were converted to number densities using atmospheric densities from ACE-FTS. Note that only the DIAL gives a direct vertical column measurement; the other instruments observe over an extended line-of-sight of up to several hundred kilometers. As seen in Figure 1, ACE-FTS partial columns are very close to either the ozonesonde or DIAL values for six of the nine days for which ACE-FTS data was available. ACE-MAESTRO partial columns are lower than those of ACE-FTS, but are within the combined error bars for all but two days.

[13] Figure 2 presents the total O₃ columns measured by the campaign FTSs and UV/visible spectrometers, along with total columns derived by combining ACE-FTS data above, and ozonesonde data below, 8.8 km (where no ACE-FTS data was available). Here the ACE-FTS total columns are within the error bars of the DOAS measurements on all but one day. Three of the four SPS measurements and the MAESTRO-G measurement agree very well with those of the DOAS. The derived ACE-FTS total columns agree well with PARIS-IR and the DOAS, while the MSC-FTS values are consistently lower.

[14] To compare the ACE-FTS O_3 profiles with those from the ozonesondes, DIAL and ACE-MAESTRO, aver-

🗉 (b)

35 35 CE FTS (4) height [km 30 30 MAESTRO VIS (4 25 25 SONDE (4) 20 20 15 15 10 10 3 5 -30-20-10 0 10 20 30 0 4 difference relative to ACE-FTS [%] O₃ mixing ratio [ppm]

Figure 3. (a) Mean O_3 profiles measured by ACE-FTS, the ACE-MAESTRO visible spectrophotometer, ozonesondes and the DIAL, using four profiles to calculate the mean, and (b) the mean of the differences relative to the ACE-FTS profile: (ACE-FTS – x)/ACE-FTS.



Figure 4. Total NO₂ columns measured by the groundbased instruments and partial columns (13.5-41.5 km) measured by the ACE instruments.

age profiles were calculated for the four days during the intensive phase of the campaign for which ACE and campaign profiles were taken within 10 hours of each other. These average profiles and the mean of the individual profile differences relative to the ACE-FTS profile are shown in Figure 3. All profiles were interpolated onto the ACE-FTS 1-km grid and no smoothing was applied to the higher vertical resolution data. The average O₃ profiles retrieved by ACE-FTS and the ACE-MAESTRO visible spectrophotometer agree to within 20% between 10 and 30 km. The average ACE-FTS O_3 profile is as much as 15% lower than the average ozonesonde profile between 15 and 30 km and by as much as 20% between 10 and 15 km. It differs by up to 10% from the average DIAL profile between 10 and 35 km. The ACE-MAESTRO visible O₃ profiles are also generally lower than the DIAL and the ozonesonde profiles.

[15] Total O₃ column maps from the World Ozone and Ultraviolet Radiation Data Centre [http://www.woudc.org/] show that the airmasses sampled by ACE did not exhibit any significant O₃ gradients and differed by less than 25 DU from the O₃ column above Eureka. The ozonesondes stayed within 33 km of the launch site and they effectively



Figure 5. (a) Mean NO₂ profiles measured by ACE-FTS and the ACE-MAESTRO UV spectrophotometer using seven profiles to calculate the means, and (b) the mean of the differences relative to the ACE-FTS profile: (ACE-FTS - x)/ACE-FTS.

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measured the same airmass as the DIAL despite an average time difference of 8 hours.

3.2. NO₂

[16] NO₂ total columns measured by the ground-based instruments show a gradual increase throughout the campaign as seen in Figure 4. This is expected, as NO₂ is known to increase slowly as the reservoir species N₂O₅ is photolyzed from the time that sunlight returns (21 February) until the sun is continuously above the horizon (14 April). After that, NO₂ reaches a steady state.

[17] The ACE overpasses were between 15:00 local time (LT) and 17:42 LT, corresponding to ground-based SZAs of 103.8° and 93.0°. The sunrise and sunset DOAS measurements were made at SZA = 90° , while the PARIS-IR and MSC-FTS spectra were recorded at SZAs between 84° and 90°. The ACE-FTS partial columns (13.5-41.5 km) fall between the DOAS sunrise and sunset total columns and within the error bars. The tropospheric contribution of NO₂ to the total column is estimated to be lower than 10% [World Meteorological Organization, 2003]. The ACE-MAESTRO partial columns are consistently higher than those of ACE-FTS. The NO₂ total columns retrieved from the PARIS-IR data are within the combined errorbars of the MSC-FTS total columns for all days. The differences between the campaign FTS and UV/visible instruments may be due to the column measurements being preferentially weighted to different altitudes for each technique.

[18] Mean NO₂ profiles calculated using seven ACE-FTS and seven ACE-MAESTRO UV spectrophotometer profiles are compared in Figure 5. The ACE-FTS and ACE-MAESTRO NO₂ profiles agree to better than 40% between 17 km and 40 km, which is within the standard deviations. The ACE-FTS results show less NO₂ than ACE-MAESTRO below 30 km but have more NO₂ above 30 km.

3.3. Temperature

[19] Temperature results from ACE-FTS, the DIAL and radiosondes were compared for the same four days as were used for O_3 comparisons. The resulting average temperature profiles (Figure 6) show differences of less than 2.5 K between ACE-FTS and the radiosondes from 10 to 30 km. Similar results are seen for ACE-FTS and the DIAL between 17 and 37 km. Above 37 km, they agree to within 1 K. The sonde and DIAL agree to within 1.5 K.

4. Conclusions

[20] The 2004 Canadian Arctic ACE Validation Campaign took place at Eureka, Nunavut. From 21 February to 9 March, measurements were taken by six ground-based instruments and balloon-borne ozonesondes. During this period, there were nine ACE occultations within 200 km of Eureka. Vertical columns and profiles have been retrieved and compared to results from the ACE satellite. Most of the ACE-FTS O₃ and NO₂ columns agree with those measured by the campaign instruments to within their combined error bars. The column retrievals from PARIS-IR and the MSC-FTS are being compared more closely to investigate the observed discrepancies.

[21] The mean O_3 profiles from Version 1.0 ACE-FTS retrievals and initial ACE-MAESTRO retrievals agree to within 20% between 10 and 30 km and the respective NO₂



Figure 6. (a) Mean temperature profiles measured by ACE-FTS, the radiosondes and the DIAL using four profiles to calculate the means, and (b) their differences in K: ACE-FTS - x.

measurements agree to within 40% between 17 and 40 km, which is within the combined standard deviations for both cases. The ACE-FTS temperature profiles agree to better than 2.5 K with the radiosonde and DIAL results. Arctic conditions are highly variable making validation campaigns a challenge, but despite the difficulties the overall agreement of the ACE measurements with the ground-based data is encouraging.

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References

- Bassford, M. R., C. A. McLinden, and K. Strong (2001), Zenith-sky observations of stratospheric gases: The sensitivity of air mass factors to geophysical parameters and the influence of tropospheric clouds, J. Quant. Spectrosc. Radiat. Transfer, 68, 657–677.
- Bassford, M. R., K. Strong, C. A. McLinden, and C. T. McElroy (2005), Ground-based measurements of ozone and NO₂ during MANTRA 1998 using a new zenith-sky spectrometer, *Atmos. Ocean*, in press.
- Bernath, P. F., et al. (2005), Atmospheric Chemistry Experiment (ACE): Mission overview, *Geophys. Res. Lett.*, 32, L15S01, doi:10.1029/ 2005GL022386.
- Bird, J. C., A. I. Carswell, D. P. Donovan, T. J. Duck, S. R. Pal, J. A. Whiteway, and D. I. Wardle (1996), Stratospheric studies at the Eureka NDSC station using a Rayleigh/Raman differential absorption lidar, paper presented at XVIII Quadrennial Ozone Symposium-96, Univ. of L'Aquila, L'Aquila, Italy.
- Boone, C. D., R. Nassar, K. A. Walker, Y. Rochon, S. D. McLeod, and P. F. Bernath (2005), Retrievals for the Atmospheric Chemistry Experiment Fourier Transform Spectrometer, *Appl. Opt.*, in press.
- Davies, J., D. W. Tarasick, C. T. McElroy, J. B. Kerr, P. F. Fogal, and V. Savastiouk (2000), Evaluation of ECC ozonesonde preparation methods from laboratory tests and field comparisons during MANTRA, in *Proceedings of the Quadrennial Ozone Symposium*, edited by R. D. Bojkov and S. Kazuo, pp. 137–138, Hokkaido Univ., Sapporo, Japan.
- Donovan, D. P., et al. (1997), Ozone, column ClO, and PSC measurements made at the NDSC Eureka Observatory (80N, 86W) during the spring of 1997, *Geophys. Res. Lett.*, 24, 2709–2712.
- Harvey, V. L., and M. H. Hitchman (1996), A climatology of the Aleutian high, J. Atmos. Sci., 53, 2088-2101.

- Kurylo, M. J. (1991), Network for the detection of stratospheric change, Proc. Soc. Photo Opt. Instrum. Eng., 1491, 169–174.
- McElroy, C. T. (1995), A spectroradiometer for the measurement of direct and scattered solar spectral irradiance from on-board the NASA ER-2 high-altitude research aircraft, *Geophys. Res. Lett.*, 22, 1361–1364.
- McLinden, C. A., J. C. McConnell, E. Griffioen, and C. T. McElroy (2002), A vector radiative-transfer model for the Odin/OSIRIS project, *Can. J. Phys.*, 80, 375–393.
- Murphy, C., et al. (2001), Validation of NDSC measurements of ozone, reservoir compounds and dynamical tracers: Results of a series of sideby-side instrument intercomparisons, paper presented at NDSC 2001 Symposium, Eur. Comm., Arcachon, France.
- Rinsland, C. P., M. A. H. Smith, P. L. Rinsland, A. Goldman, J. W. Brault, and G. M. Stokes (1982), Ground-based infrared spectroscopic measurements of atmospheric hydrogen cyanide, *J. Geophys. Res.*, 87, 11,119– 11,125.
- Rinsland, C. P., et al. (1998), Northern and Southern Hemisphere groundbased infrared spectroscopic measurements of tropospheric carbon monoxide and ethane, *J. Geophys. Res.*, 103, 28,197–28,217.
 World Meteorological Organization (2003), Scientific assessment of ozone
- World Meteorological Organization (2003), Scientific assessment of ozone depletion: 2002, Global Ozone Res. Monit. Proj. Rep. 47, Geneva.
- J. R. Drummond, A. Fraser, T. E. Kerzenmacher, K. MacQuarrie, C. T. McElroy, C. Midwinter, K. Strong, J. Walker, and H. Wu, Department of Physics, University of Toronto, 60 St. George Street, Toronto, Canada M5S1A7. (tobias@atmosp.physics.utoronto.ca)

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