Probing Atmospheric Composition over Canada using Ground-based FTIR Spectroscopy

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Abstract: We describe the use of ground-based Fourier transform (FTIR) solar absorption spectroscopy to measure vertical columns of tropospheric and stratospheric trace gases. We focus on measurements made at the University of Toronto Atmospheric Observatory and the Polar Environment Atmospheric Research Laboratory in the high Arctic, and introduce the new Canadian FTIR Observing Network.

OCIS codes: (010.1280) Atmospheric composition; (010.1120) Air pollution monitoring; (300.6340) Spectroscopy, infrared; (300.6300) Spectroscopy, Fourier transforms; (010.0280) Remote sensing and sensors

1. Introduction

Infrared spectroscopy provides a powerful technique for detecting trace gases and has been used to measure atmospheric composition from the ground, balloons, and aircraft since the 1960s. Fourier transform infrared (FTIR) spectrometers offer high spectral resolution, wide spectral coverage, and excellent throughput, which enable the measurement of many species simultaneously. At about two dozen sites worldwide, FTIR spectrometers are used to retrieve vertical columns and some vertical profile information using solar infrared absorption spectroscopy under the auspices of the Network for the Detection of Atmospheric Composition Change (NDACC) [1¸2], the Total Carbon Column Observing Network (TCCON) [3], and the MUlti-platform remote Sensing of Isotopologues for investigating the Cycle of Atmospheric water (MUSICA) network [4]. In Canada, there are currently two stations having FTIR spectrometers affiliated with these networks (Toronto and Eureka), with seven more instruments in use or available for deployment; all of these are being used to create a new Canadian FTIR Observing Network (CAFTON). This paper provides a brief overview of the Toronto and Eureka measurements and science highlights, and introduces CAFTON.

2. FTIR measurements at the University of Toronto Atmospheric Observatory

The University of Toronto Atmospheric Observatory (TAO) was commissioned in 2001, and an ABB Bomem DA8 FTIR spectrometer was installed as its primary instrument, subsequently joining NDACC [5]. Since 2002, solar absorption spectra have been recorded routinely with InSb and MCT detectors, using narrow band optical filters over a spectral range of 720 to 4300 cm^{-1} at a resolution of 0.004 cm^{-1}. Using the SFIT2 optimal estimation retrieval algorithm [6], total and partial columns of O_3, HF, HCl, NO, NO_2, N_2O, CO, CH_4, C_2H_6, and HCN have been measured. A ten-year time series of TAO trace gas measurements has now been obtained (see Figure 1), and has been compared with data from the DA8 at Environment Canada's Centre for Atmospheric Research Experiments (CARE) at Egbert, 60 km north of Toronto. The combined TAO/CARE dataset of HF, HCl, N_2O, and O_3 columns has been used to identify polar vortex intrusion events over Toronto, and establish a dynamical cause for some of the winter/spring variability of stratospheric trace gases observed at this mid-latitude site [7]. In another study, the GEOS-Chem 3-D chemical transport model has been used to interpret the FTIR measurements of tropospheric O_3, CO, and C_2H_6, and to identify the sources of air pollution over Toronto.

3. FTIR measurements at the Polar Environment Atmospheric Research Laboratory

The Polar Environment Atmospheric Research Laboratory (PEARL) is located in the Canadian high Arctic at Eureka, Nunavut (80°N) and was established by the Canadian Network for the Detection of Atmospheric Change (CANDAC) in 2005. PEARL houses several instruments that measure chemical constituents, including a Bruker IFS 125HR FTIR spectrometer (on site since 2006) [8], which replaced the former DA8 FTIR spectrometer (operational from 1993-2008) [9]. The DA8 was an NDACC instrument, while the 125HR has both mid-infrared and near-infrared measurement capabilities, and is affiliated with NDACC, TCCON, and MUSICA. The first five years of 125HR measurements are shown in Figure 2.

The combined DA8/125HR dataset has been used to investigate chemical ozone loss above Eureka, particularly in winter/spring 2011, when the polar vortex was typically circular, cold and centered above the pole, with Eureka mostly inside the vortex from October 2010 until late March 2011 [10]. Unusually low ozone, HCl, and HNO_3 total columns were observed in spring 2011 in comparison to the previous 14 years. The 2011 FTIR measurements were consistent with the low temperatures and polar stratospheric clouds observed over Eureka. The PEARL 125HR also measures tropospheric species, including CO, HCN, C_2H_6, C_2H_2 and C_2H_4, which show periodic enhancements due to the transport of smoke plumes from biomass burning events [11]. These are correlated with simultaneous and co-located measurements of the 500-nm fine mode component of the aerosol optical depth from AERONET/AEROCAN CIMEL sunphotometers. The trace gas columns have been converted into emission ratios relative to CO, and used to derive emission factors for boreal forest fire events.

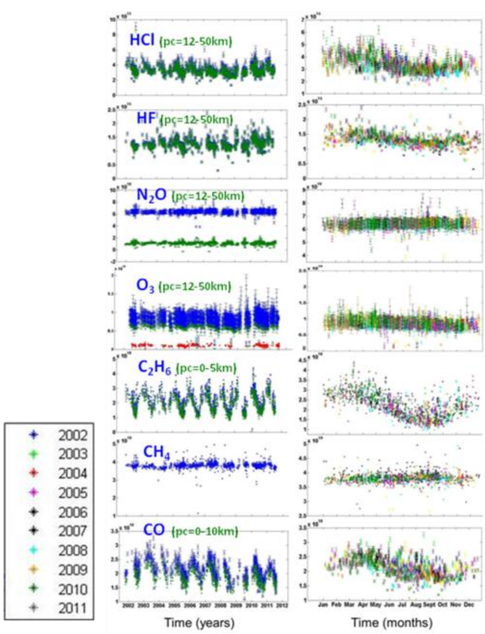

Figure 1: Ten years of TAO DA8 FTIR measurements at Toronto. The panels on the left show the time series of total columns (blue), partial columns with altitude ranges as indicated (green), and 0-12 km partial columns for ozone (red). The panels on the right combine all of the measurements, colour-coded by year, to illustrate the seasonal cycles.

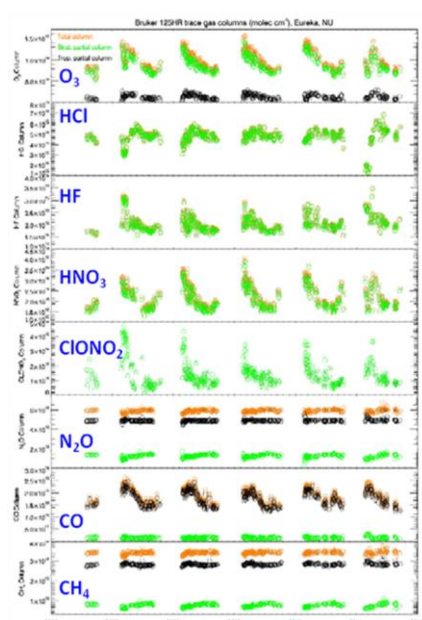

Figure 2: The first five years of PEARL 125HR FTIR measurements at Eureka. The time series of 0-100 km total columns (orange), 10-50 km stratospheric partial columns (green), and 0-10 km tropospheric partial columns (black) are shown.

4. The Canadian FTIR Observing Network

In addition to the FTIR spectrometers mentioned above, there is a DA8 operating at the Dalhousie Atmospheric Observatory in Halifax, an ABB Bomem Extended-range Atmospheric Emitted Radiance Interferometer (E-AERI) at PEARL [12], and the ABB Bomem Portable Atmospheric Research Interferometric Spectrometer for the Infrared (PARIS-IR) for deployment on field campaigns [13,14]. While the E-AERI is a moderate-resolution (1 cm^{-1}) instrument, it measures the absolute downwelling infrared spectral radiance, and thus runs year-round, including the four months of polar night when the solar-viewing spectrometers at PEARL are not operated. E-AERI spectra provide information about radiative balance, trace gases, and cloud properties in the high Arctic.

In a new initiative, the nine ground-based Canadian FTIR spectrometers currently making, or capable of making, atmospheric measurements are being coordinated through the creation of the Canadian FTIR Observing Network. This includes the three DA8 instruments at Toronto, Halifax, and Egbert; the 125HR and E-AERI at Eureka; the deployment of a refurbished DA8 and a new 125HR at a new site in western Canada; and two mobile instruments, a new Bruker IFS 125M and PARIS-IR. CAFTON will involve: (i) coordination and enhancement of the measurement capabilities of the nine FTIR spectrometers, (ii) implementation of new and improved methods for measuring atmospheric constituents, and (iii) use of the GEOS-Chem global chemical transport model [15] and the STILT regional particle dispersion model [16] to investigate processes related to the origin, concentration, and transport of airborne contaminants. The resulting network will provide ongoing measurements of the atmosphere over Canada. The stations of the network will also be anchor points, providing well-characterized data sets that can be used to validate measurements made by current and future satellite missions.

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