Trace gas retrievals at two Arctic sites using emission spectroscopy: Is the atmospheric composition different between Eureka, Nunavut (80°N) and Barrow, Alaska (71°N)?

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1. INTRODUCTION

The Arctic experiences prolonged periods of total darkness in the winter and continuous daylight in the summer, influencing the atmosphere and its composition in ways that are still not fully understood. The Canadian Network for the Detection of Atmospheric Change has equipped the Polar Environment Atmospheric Research Laboratory (PEARL, Eureka, Nunavut, Canada, 80°05'N, 86°42'W) with several spectrometers that measure atmospheric composition. Similarly the U.S. Department of Energy has established numerous ground-based observatory facilities around the world within the Atmospheric Radiation Measurement (ARM) Program, with one located at the North Slope of Alaska (NSA), Barrow, Alaska.

At both sites, an Extended-range Atmospheric Emitted Radiance Interferometer (E-AERI), a ground-based Fourier transform infrared (FTIR) spectrometer, is used to measure the absolute downwelling infrared emission from the atmosphere between 400 and 3000 cm^{-1}. A number of trace gases have emission features in this region, enabling simultaneous measurements of their total column abundances. The E-AERI has a moderate resolution of 1 cm^{-1} and a high sensitivity to the lower troposphere to provide information about trace gases budgets.

Figure 1: Arctic map with location of Barrow, AK (blue star) and Eureka, NU (pink star).

At PEARL, the instrument was installed in October 2008. A similar instrument, the University of Idaho's Polar AERI (P-AERI) was installed at PEARL from March 2006 to June 2009. Measurements are taken year-round, including polar night when the solar-viewing spectrometers at PEARL are not operated. At NSA, the instrument has been operating since February 1998. Both datasets can be used to fill in the measurement gap during the Arctic night-time period when less is known about atmospheric composition.

Total columns of CO, CH_4, N_2O and O_3 have been retrieved from 2006 to 2014 (except in 2010) at PEARL and, from 2000 to 2014 at NSA using the SFIT4 algorithm including the emission add-on. These measurements are used to investigate the annual, seasonal and diurnal variabilities of these four species in the high Arctic at two different sites.

2. METHOD

E-AERI = Extended-range Atmospheric Emitted Radiance Interferometer

- Infrared Fourier Transform Spectrometer (FTS) with 1cm^{-1} resolution
- Measurements of accurately calibrated downwelling infrared thermal emission from the atmosphere
- Extended wavelength range covers **400**-3000 cm^{-1} (3-25 µm) to investigate the IR surface cooling in the Arctic
- **High sensitivity to tropospheric trace gases**
- **Measurements are independent of sunlight**

Retrievals of trace gases using SFIT4 algorithm, which include the emission add-on to calculate the full radiative transfer.

To be comparable to previous work using SFIT2 retrieval algorithm, all the parameters and settings (input files) in SFIT4 are identical to those used in SFIT2.

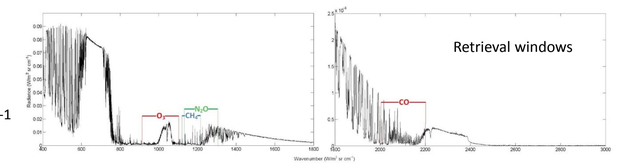

Figure 2: Retrievals windows for each trace gas.

Target species	Wavenumber range (cm-1)	Miniwindow size (cm-1)	Interfering species
CO	2000 – 2200	200	O_3, N_2O, H_2O, OCS
CH_4	1150 – 1229	79	SO_2, HDO, H_2O, O_3, CCL_2F_2, HNO_3, CH_3D, N_2O
N_2O	1160 – 1300	140	CH_4, SO_2, HDO, H_2O, O_3, CCL_2F_2, HNO_3, CH_3D
O_3	950 – 1100	150	H_2O, CO_2, O_3^{676}, O_3^{667}, O_3^{686}, O_3^{668}

Table 1: Summary of the specifications of target species.

3. TIMESERIES AT EUREKA AND BARROW

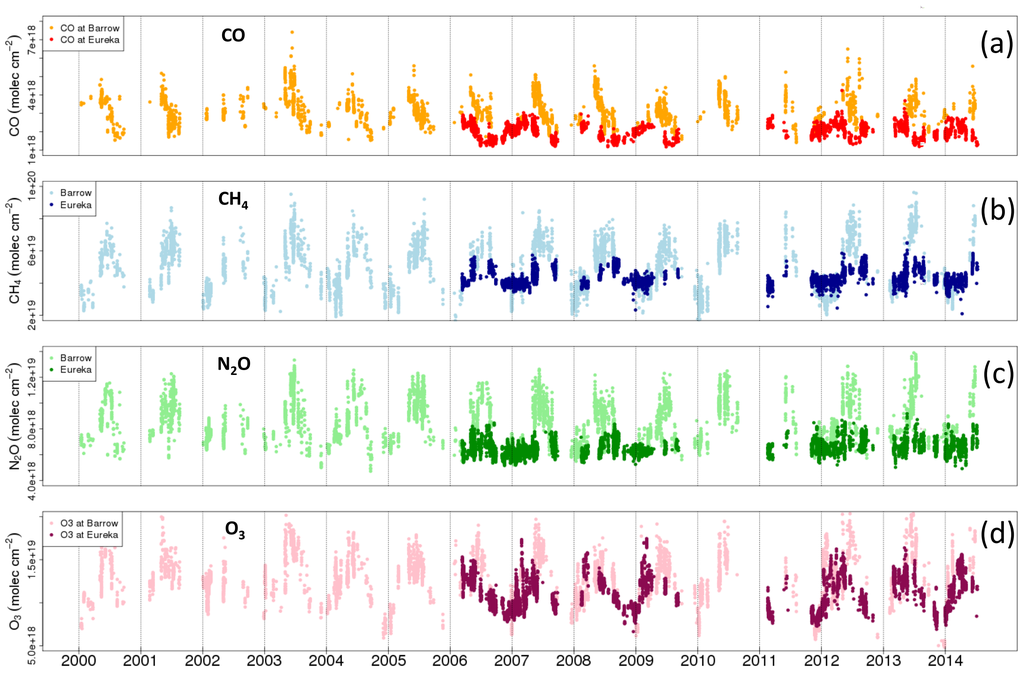

Figure 3: Time-series of zenith column densities (in molec cm^{-2}) of carbon monoxide (a), methane (b), nitrous oxide(c) and ozone (d) at Barrow, AK from 2000 to 2014 (light colors) and Eureka, NU from 2006 to 2014 (dark colors).

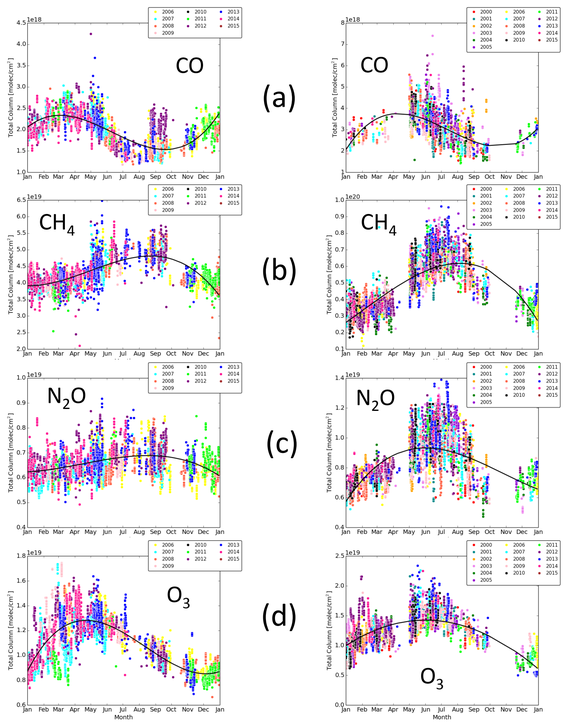

Figure 4: Seasonal cycle of column densities (in molec cm^{-2}) of CO (a), CH_4 (b), N_2O (c) and O_3 (d) for Eureka, NU on the left side panel and for Barrow, AK on the right side.

- Clear seasonal cycle for carbon monoxide and ozone at both sites, with an accumulation of these trace gases during the winter and a decrease during the summer due to the photochemistry.
- Gaps in datasets are mostly due to cloudy days and non-convergent retrievals.
- Higher concentrations of these four trace gases at Barrow during the summer months, from May to October, and similarly, observations of higher concentrations of CH_4 and N_2O at Eureka for the same period → **Artefact due to water vapour spectral line mis-fitting?**

4. INVESTIGATION OF THE "CH_4 SUMMER ARTEFACT":

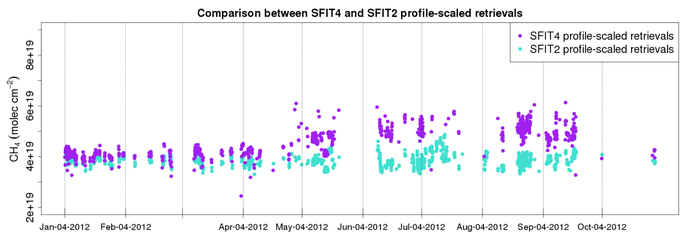

Figure 5: Methane column densities measured at Eureka in 2012 using SFIT2 (in turquoise) and using SFIT4 (in purple).

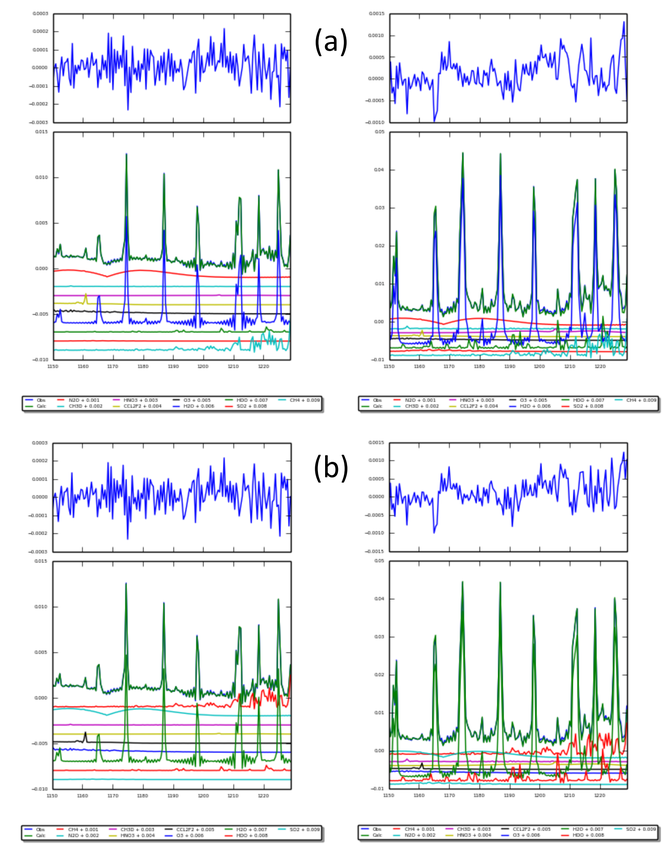

Figure 6: Sample of a winter (Jan 14, 2012; left figure) and a summer (Aug 24, 2012; right figure) spectral fits for retrievals of CH_4 at Eureka. The upper panel shows scaled-profile retrieval (a) and the lower panel corresponds to profile retrieval (b). Inside each panel, the upper window represents the residual (in W/[m^2 $srcm^{-1}$]) between the measured and the calculated spectra. The bottom window shows the measured (blue) and the fitted spectra (green), and the interfering species in different colors.

Figure 5 shows the column densities of methane retrieved using SFIT4 compared to retrievals using SFIT2. We can clearly see a "step" in the CH_4 concentrations retrieved using SFIT4 between May and October 2012. This summer feature appears consistently for CH_4 and N_2O total columns retrievals for Eureka and for all four traces gases retrieved at Barrow.

The residual of the summer spectrum (Fig. 6) is larger than the winter one. It is likely due to the water vapour line which is stronger in summer.

5. POLAR NIGHT AND SPRING

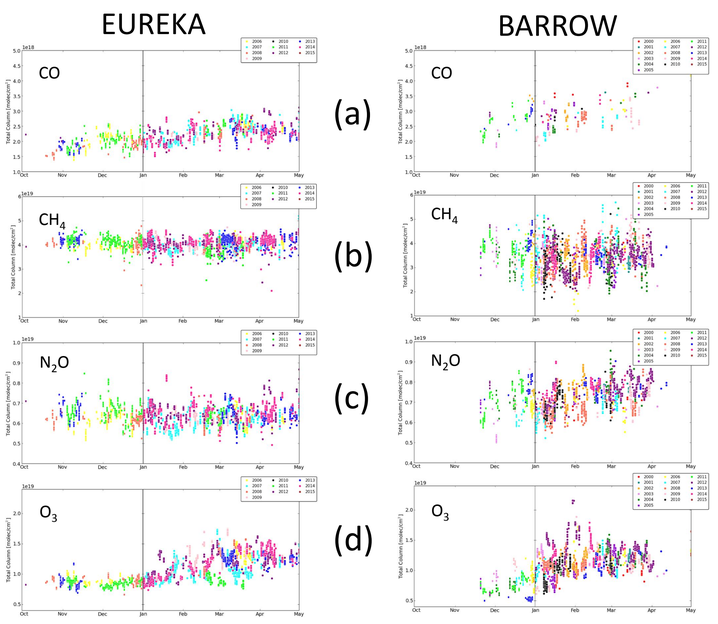

Figure 7: Column densities of CO (a), CH_4 (b), N_2O (c) and O_3 (d) at Eureka, NU on the left and at Barrow, AK on the right side.

Carbon monoxide in the winter:
- Accumulation of CO during the winter, with a maximum in March
- Sparse CO column densities obtained at Barrow due to cloudy days and non-convergent retrievals
- Higher values of CO at Barrow compared to Eureka

Methane and **Nitrous Oxide**:
- Flat trends for CH_4 and N_2O: No accumulation during the winter, concentrations stable except for N_2O in Barrow
  → Slight increase in spring?
- Higher amplitudes for Barrow

Ozone in the polar night:
- Accumulation of ozone during the polar night at both sites
- Identification of the ozone depletion in 2011 over Eureka in February and March (Manney et al., 2011; Adams et al., 2012; Lindenmaier et al., 2012)
- Ozone depletion observed in December 2013 at Barrow
- Evidence of ozone enhancements in January and February 2012 at both sites. Total ozone maps and air masses back-trajectories indicate that it was clearly a stratospheric ozone contribution.

6. CONCLUSION

1) What is new? → First trace gas retrievals from AERI instruments:
   - from 2000 up to now at Barrow, Alaska
   - from 2006 to now at Eureka, Nunavut

   → The long-term time-series will help us to better understand the changes in the atmospheric composition of the Arctic region and document the atmospheric composition during the polar night.

2) Is the atmospheric composition different between Eureka and Barrow? → YES, in term of concentrations

3) How different is it?
   - Higher trace gases concentrations in Barrow and Eureka (for CH_4 and N_2O), called here the ''summer feature'' → **Need further investigation**
   - Higher ozone total column in January: Identification of stratospheric contribution to the tropospheric ozone budget during the winter
   - Lower ozone total columns during the winter: Possible ozone depletion events

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