Airborne DOAS measurements of aerosol extinction and NO_2 profiles in Arctic: two cases studies and their transport interpretation

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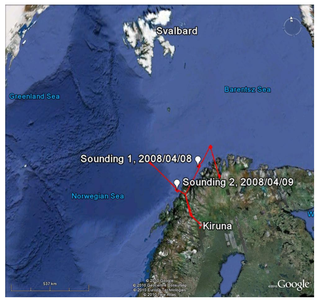

Fig.1 Tracks of the flights as0831 (8 April 2008) and as0833 (9 April 2008)

1.Context and motivations

The CNRS-spring campaign was based in Kiruna, Sweden and took place between 30 March and 14 April 2008. One scientific objective was the study of pollution long-range transport to Arctic. The ATR-42 from SAFIRE performed 12 flights above the Norway Sea. We present here measurements of the flights of 8 and 9 April 2008, during which the aircraft performed vertical soundings of the troposphere (c.f. Fig.1), respectively at 71°N, 22°E and 70°N, 17.8°E. Our goal was to retrieve aerosol and trace gases profiles from a DOAS instrument installed

2. A new instrument: the Airborne Limb Scanning DOAS(ALS-DOAS)

Developed at our institute, the ALS-DOAS records scattered-light-spectra in a near-limb geometry, through a BK-7 port on the ATR-42 (see Fig.2). The telescope is mounted on a stepper motor axis and scans between –5 and 5° to maximize the information content of the measurements.

Range : 332-450nm

Resolution : 0.5 nm

Field of view: 1°

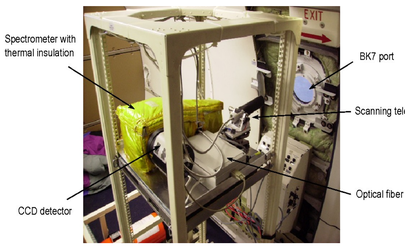

Fig2. The ALS-DOAS instrument onboard the Safire

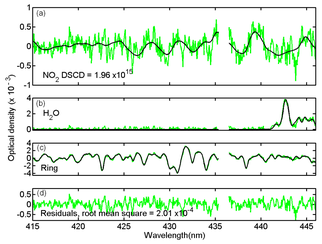

Fig.3 Exermple of NO_2 DOAS fit

3. Methods

a)DOAS analysis

Spectra are evaluated using the DOAS technique, yielding the integrated absorption along the line-of-sight compared to a reference spectrum, namely the differential slant column density (DSCD).

Fittting windows:

-[340nm 370nm] for O_4

-[415nm 446nm] for NO_2

3. Methods

b) Radiative transfer modeling and weighting functions

To interpret the O_4 and NO_2 DSCD measurements, it is necessary to assess the light path in the atmosphere. We use the radiative transfer code UV-Spec DISORT, with a model atmosphere built from in-situ aircraft measurements of temperature, pressure, and ozone. Simulations with the radiative transfer code reproduce qualitatively the DSCD measurements during the sounding (Fig.4). This represents the measurement forward model.

The contribution to the measuremed ***DSCD*** of a given parameter ***x*** in each layer ***i*** is quantified with the weighting functions, calculated with the radiative transfer model, as the partial derivative: $\frac{\partial DSCD}{\partial x_i}$

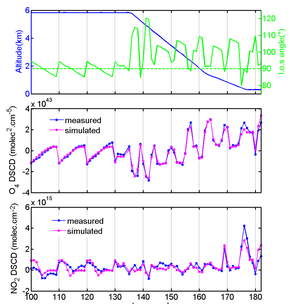

Fig.4 Measured NO_2 and O_4 DSCD versus radiative transfer simulations for the sounding 1

In our case, we retrieve the aerosol extinction and the NO_2 concentration, respectively from the O_4 and NO_2 DSCD measurements. Those quantities being positive, we also calculate the weighting functions compared to the logarithm of these quantities to constrain positively the retrievals. Some of the weighting functions relative to the NO_2 concentration in linear and logarithmic scale are shown in Fig.5, they were calculated by perturbations on the layers extinction and NO_2 concentration. They correspond to the sounding 1 (8 April 2008). The maximum sensitivity is close to the observation altitude due to the limb geometry, wich makes this kind of measurements particularly interesting to study the free troposphere. Ground-based and satellite measurements are generally poorly sensitive to this altitude

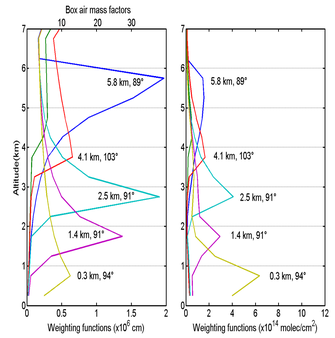

Fig.5 Weighting functions relative to the NO2 concentration for some measurements of the sounding 1 in linear(left) and logarithmic scale(right).

3. Methods

c) Inversion scheme

We use the maximum a posteriori solution to retrieve the profiles, assuming gaussian statistics for the observation error (S_e) and the a priori (x_a,S_a). Due to the non-linearity of the problem, iterations are necessary to reach the solution:

$$x_{i+1} = x_i+(S_a^{-1}+K_i^T S_e^{-1} K_i)^{-1}[K_i^T S_e^{-1} (y-F(x_i))-S_a^{-1}(x_i-x_a)]$$

Where K_i is the weighting functions matrix, y the DSCD series and F the forward model. The solution is characterised (resolution, weight of the measurements relative to the apriori) by the averaging kernels matrix A:

$$A = (K_i^T S_e^{-1} K_i+S_a^{-1})^{-1} K_i^T S_e^{-1} K_i$$

4. Results and interpretations

a) Sounding 1 (8 April 2008)

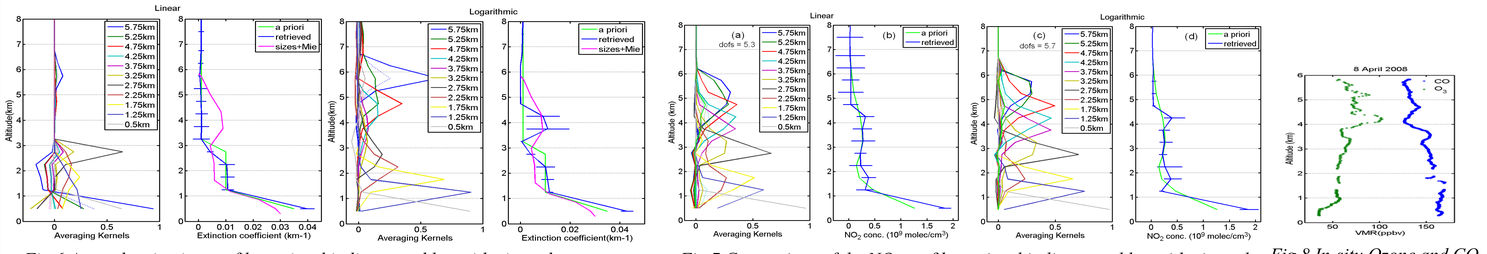

Fig.6 Aerosol extinction profile retrieval in linear and logarithmic scale

Fig.7 Comparison of the NO_2 profile retrieval in linear and logarithmic scale

Fig.8 In-situ Ozone and CO

We show on Fig.6 and 7 the retrievals for the sounding 1. In the boundary layer, enhanced NO2 ($1.9x10^9$ molec/cm^3) and extinction(0.04 km^{-1}) are found with the two inversion approaches. In the free troposphere, a layer with a lower extinction (0.01 km^{-1}) is only seen with the logarithmic approach, and confirmed with in-situ data.Backtrajectories and retroplumes performed with Hysplit (Fig. 11) and Flexpart(Fig. 9) allows to attribute a local/regional origin for the boundary layer air, whereas the air sampled in the free troposphere seems to be a mixed between stratospheric and tropospheric air. This is coherent with the ozone and CO in-situ data(Fig 8).

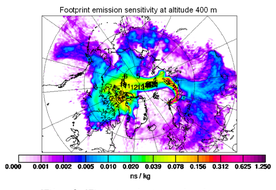

Fig.9 Footprint emission sensitivity for the air in the boundary layer (sounding 1)

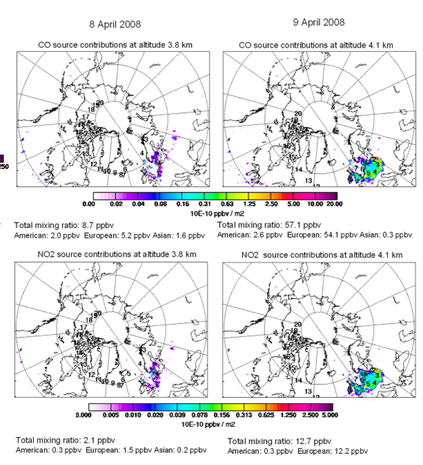

Fig. 13. CO and NO_2 source contributions for the altitude with maximum CO concentration in the two soundings.

b) Sounding 2 (9 April 2008)

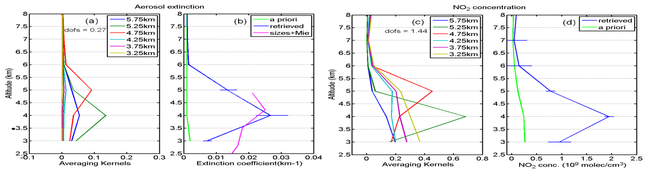

Fig.10 Retrievals of aerosol extinction(left) and NO_2(right) for the sounding 2

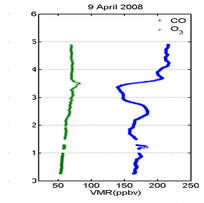

Fig.12 In-situ Ozone and CO

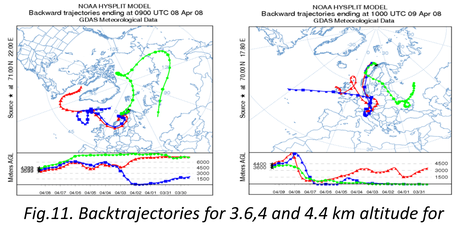

Fig.11. Backtrajectories for 3.6,4 and 4.4 km altitude for flight 1(left) and flight 2 (right)

Fig 10 presents the retrieved profiles for souding 2. Due to clouds, the profile's lowest altitude is 3 km. A layer with enhanced extinction(0.025 km^{-1}) corresponds to an increase in NO_2 ($1.95x10^9$ molec/cm^3) at 4 km. In situ data also show a CO layer from 4 km altitude. Backtrajectories (Fig. 11) and source contributions (Fig 13) indicate a central european origin for the considered air masses, which is coherent with the pollution detected.

Reference and aknowledgements

Merlaud et al. *Airborne DOAS measurements in Arctic: vertical distributions of aerosol extinction coefficient and NO_2 concentration,* submitted to ACPD

This work was supported by the Belgian Federal Science Policy (contract MO/35/028). POLARCAT-France was funded by French ANR, CNES, CNRS-INSU (LEFE-CHAT), IPEV and EUFAR, the latter funded also our participation to the campaign and a couple of additional flight hours.We are grateful to the UMS SAFIRE for their support in the instrument installation and providing the aircraft data