Ground-based CO observations at the Jungfraujoch: Comparison between FTIR and NDIR measurements

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Introduction

While the usage of space-borne instruments has become increasingly important for measuring the atmospheric composition on a global scale, ground-based measurements are still crucial. Ground-based measurements often provide highly accurate data with high time resolution, ideally suited for validation purposes. Here we present preliminary results from an inter-comparison of two CO time series (1997-2004) observed at the Jungfraujoch with in situ Non-Dispersive Infrared (NDIR) technique and remote sensing Fourier Transform Infrared (FTIR) solar absorption spectrometry.

Fig 1. Location of the Jungfraujoch site

The Jungfraujoch Station

The Jungfraujoch station (46.5°N, 8°E, 3580 m above sea level) is located on a mountain saddle between the Jungfrau (4158 m asl) and Mönch (4099 m asl) peaks in the Swiss Alps. Due to its unique place, the year-round accessibility, and the excellent infrastructure, the Jungfraujoch research station is well suited for long-term ground-based in-situ as well as remote sensing monitoring of trace gases with only little influence of pollution from the atmospheric boundary layer.

In situ NDIR technique

- Operated by Empa
- Cross Flow Modulated NDIR Absorption technology (Horiba APMA-360)
- Uses a built-in IR light source and samples in situ ambient air
- Sample gas and reference gas are injected alternately into the measurement cell. Sample air is taken to generate CO-free reference gas by using a catalyst to oxidise CO to CO2.
- Data are 10 minutes averages of the local CO concentration at 3580 m asl.
- The overall measurement uncertainty for these 10 minutes mean values is estimated to be <10% below 100 ppbv and <5% above 100 ppbv.

Remote sensing FTIR solar absorption spectrometry

- Performed by the University of Liège (ULg) in collaboration with BIRA-IASB
- FTIR detects the downwelling direct solar radiation.
  - requires clear sky conditions!
- Retrieval of CO profile: SFIT2 algorithm + HITRAN2004
- CO a priori profile = a-priori ATMOS version 3 occultation measurements performed in Northern mid-latitudes in November 1994
- Retrieved profile has a limited vertical resolution (typical DOF ~ 2.18)
- Information content is determined by the Averaging Kernels [Rodgers, 2000]. The retrieved volume mixing ratio (vmr) profile xr is thus related to the true profile x and to the a priori profile xa by :

$$xr = xa + A(x - xa) + (\text{errors})$$

in which A is the matrix whose rows are the averaging kernels.

- Taking the lowest retrieved layer would depend too much on the a priori information

⇒Take average vmr over the lowest 4 layers (from 3.58 up to 7.0 km) !

- The DOF of this merged layer is >1 and any impact of the a-priori is negligible.
- The random error on this merged layer is estimated to be 7%, calculated from a typical retrieval error covariance matrix.

NDIR-FTIR inter-comparison

First, we have compared the FTIR and NDIR data sets as such (Fig. 2a).

It is clear that NDIR data exhibits far more scatter than FTIR, which can be readily attributed to the sampled airmass, i.e. in situ vs. aggregated signals from 3.58 to 7 km (Note that similar resolution losses apply to satellite data, limiting its role as Air-Quality monitors).

The overall trends over the 1997-2004 period are different for FTIR and NDIR!

All data (in ppb/year)

FTIR: -0.93 ± 0.12 NDIR:-4.7 ±0.01

To eliminate (as much as possible) the impact of the fact that FTIR measures clear sky data only, only data points taken within 6 minutes of each other are kept (fig 2b). This temporal overlap criterion has been chosen very strict taking into account the short-term variability especially of the in-situ CO observations.

The mean diurnal variability of CO (max-min NDIR CO for each day, using only days which feature a FTIR measurement) is 67 ± 37 ppb (see Fig. 3).

After this data selection the trend differences remain almost the same (see Fig. 2b).

6min overlap (in ppb/year)

FTIR: -0.99 ± 0.12 NDIR:-5.1 ±0.14

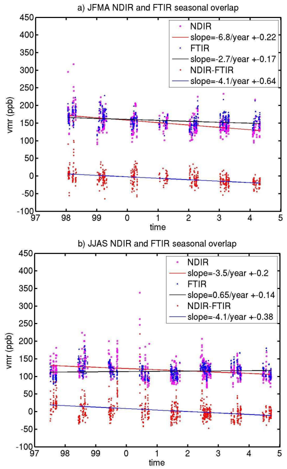

Fig. 4 Jan-Apr (a) and Jun-Sep (b) CO values and trends (in ppb/year, overlapping data only)

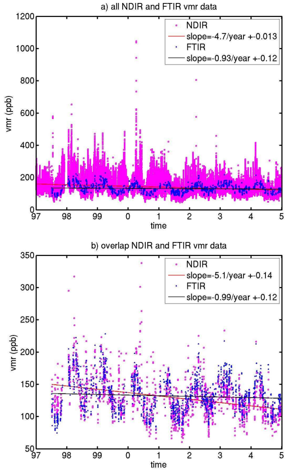

Fig. 2 All (a) and temporally overlapping (b) FTIR (3.58-7km averages) and NDIR data

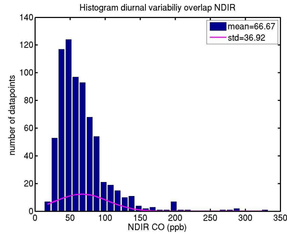

Fig. 3 Histogram of CO daily range (max-min CO) + gaussian distribution

Seasonality

CO's seasonal cycle exhibits high values during late winter, early spring (Jan-Apr), and low values during summer (Jun-Sep).

The decrease in CO is more significant during the Jan-Apr period (Fig 4.) (by ~ 3 ppb/year).

Jun-Sep FTIR CO even shows a slightly positive trend.

However the difference in trends between NDIR and FTIR remains -4.1 ppb/year for both periods

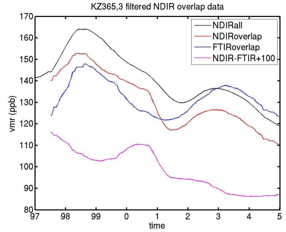

Fig. 5 KZ filtered NDIR and FTIR data (cutoff at 1.7 years) and its difference.

Free Troposphere CO

CO data points, representative for the free troposphere, are selected by rejecting data which feature a high diurnal variability (>50%) and high NOy/CO values (>0.005). Apart from a bias shift no difference in trends is observed.

Taking 10-20km FTIR data, results in a bias shift and a clear decrease in the seasonal cycle amplitude, but has no obvious impact on the long term trend.

Conclusions

- The correlation between NDIR and FTIR measurements (3.6-7.0 km) is very good (R=0.72)
- However the different trends for NDIR and FTIR data remain unclear and further research into the cause is ongoing.
- Given the difference in sampled air-mass it is not unlikely for such a difference to occur, however…
- Removing PBL contaminated air from the NDIR dataset, does not significantly impact the overall trend.
- The trend difference is gradual and features no strong seasonal dependence (unlike the trends themselves)
- FTIR window selection impacts the CO trend

Acknowledgements

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Kolmogorov-Zurbenko filter

The KZ filter is a low pass filter which separates the time series at a selected cutoff frequency. The thus obtained long term trend (cutoff frequency = 1.7 years) is shown in Fig 5. Apart from the year 2000 (which featured some unusually high NDIR data), the NDIR-FTIR seems to be gradually decreasing until mid 2003.

Apart from 1998 and 2002-2003 (large biomass burning events), the overal trend is negative.

Also clear is that from 2001 onwards, (3.56-7km averaged) FTIR CO values are in fact higher than those measured by in situ NDIR!

FTIR window selection

Retrieval from the 2050-2160 cm^{-1} window (using ^{12}CO and ^{13}CO absorption lines) [as used throughout this poster] yields a trend of -0.83 ppb/year. [only FTIR data, taken within 1/2h of a 4250 cm^{-1} measurement]

Retrievals near 4250 cm^{-1} (using ^{12}CO lines only) yield a trend of + 1.9 ppb/year.

The cause of this significant difference is under investigation.

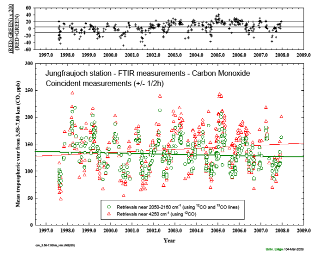

Fig. 6 Impact of µwindow selection upon the CO retrieval. Green data points are the 'standard' CO data retrieved according to the UFTIR project (http://www.nilu.no/uftir), while the red data points are derived from taking ^{12}CO lines only.

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