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Simulations of atmospheric CO_2 and $\delta^{13}C$-CO_2 compared to real-time observations at the high altitude station Jungfraujoch, Switzerland

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Overview

Motivation

Evaluating atmospheric transport simulations of greenhouse gases against their observations helps refining bottom-up estimates of their fluxes. Thereby, it helps in identifying gaps in our understanding of regional and category-specific contributions to atmospheric mole fractions. This insight is critical in the efforts to mitigate anthropogenic environmental impact. Beside total mole fractions, stable isotope ratios provide further constraints on source-sink processes.

Objectives

While the Jungfraujoch research station ("JFJ") observes European atmospheric background conditions for the majority of time owing to its high altitude, regional signals are observed when the station is influenced by air masses from the planetary boundary layer (PBL). Observation-validated simulations shall allow for an in-depth evaluation of the contribution of different CO_2 emission sources and sinks to regional CO_2 concentrations at JFJ.

Methods

We present two receptor-oriented model simulations for carbon dioxide (CO_2) mole fraction and $\delta^{13}C$-CO_2 stable isotope ratios for a nine year period, from 2009-2017, at the High Altitude Research Station Jungfraujoch "JFJ" (Switzerland, 3580 m asl). Simulations are performed at a 3-hourly time resolution.

We compare the simulations with and evaluate them against a unique data set of highly time-resolved continuous in-situ observations of CO_2 mole fractions and $\delta^{13}C$-CO_2 stable isotope ratios measured by quantum cascade laser absorption spectroscopy (QCLAS, see observations time-series in Figure 1).

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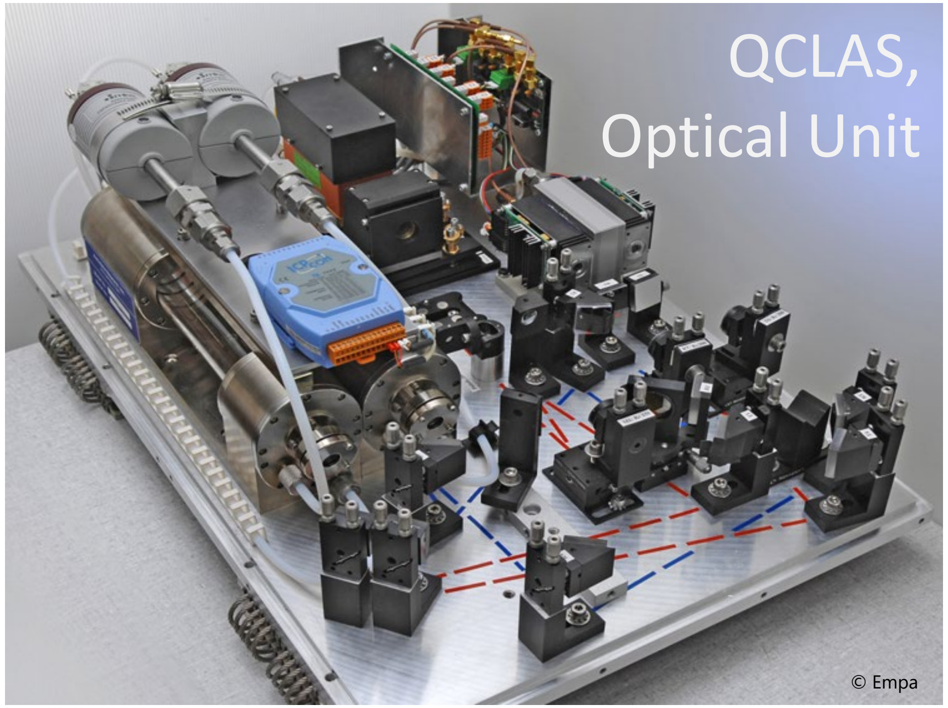
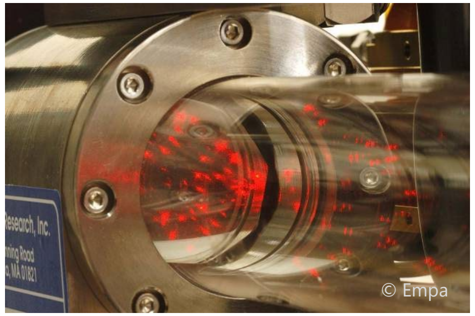
Measurement technique and measurement site





Natural Abundance:

$^{12}C^{16}O_2$ 98.42 %
$^{13}C^{16}O_2$ 1.100 %
$^{12}C^{16}O^{18}O$ 0.395 %

δ Notation:

$$\delta^{13}C = \frac{[^{13}C/^{12}C]_{Sample}}{[^{13}C/^{12}C]_{VPDB}} - 1$$

$$\delta^{18}O = \frac{[^{18}O/^{16}O]_{Sample}}{[^{18}O/^{16}O]_{VPDB}} - 1$$

VPDB = «Vienna PeeDee Belemnite» Reference Scale

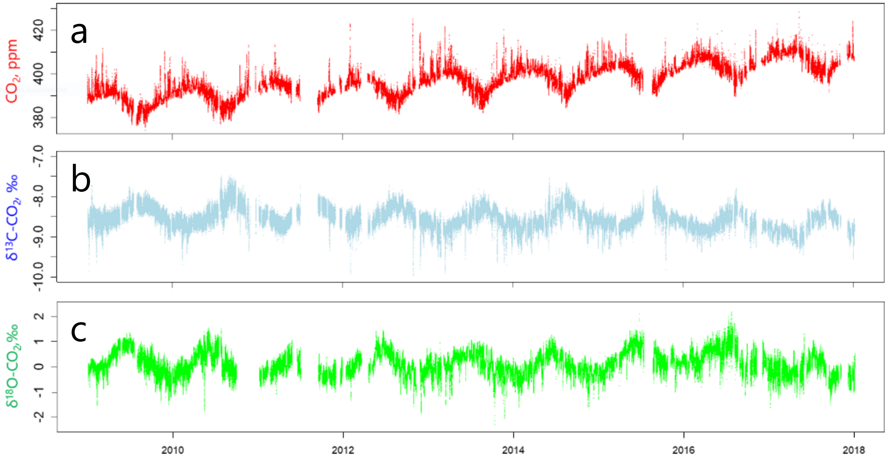


Figure 1. Atmospheric in-situ measurements of (a) CO_2 concentration (ppm), (b) $\delta^{13}C$- and (c) $\delta^{18}O$-CO_2 ratio (‰) at Jungfraujoch for 9 years (2009-2017).

Related References:
Nelson et al., Applied Physics B (2008)
Tuzson et al., Applied Physics B (2008
Tuzson et al., Infrared Physics & Technology (2008)
Sturm et al., Atmos. Meas. Techn. (2013)

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Simulations framework

Receptor-oriented CO_2 simulations

The model simulations of CO_2 were performed on a 3-hourly time-resolution with two backward Lagrangian particle dispersion models driven by two different numerical weather forecast fields: FLEXPART-COSMO and STILT-ECMWF.

Anthropogenic CO_2 fluxes were based on the EDGAR v4.3 emissions inventory aggregated into 14 source categories representing fossil and biogenic fuel uses as well as emissions from cement production.

Biospheric CO_2 fluxes representing the photo-synthetic uptake and respiration of 8 plant functional types were based on the Vegetation Photosynthesis and Respiration Model (VPRM).

Simulation of ambient $\delta^{13}C$-CO_2 ratios

The simulated CO_2 emissions per source and sink category were weighted with category-specific $\delta^{13}C$-CO_2 signatures from published experimental studies (see Table 1, Eq. 1, Figure 3).

Background CO_2 values at the boundaries of both model domains were taken from global model simulations and the corresponding $\delta^{13}C$-CO_2 values were constructed thereof, and combined with the mixed $\delta^{13}C$-CO_2 source signatures (see Eq. 2)

Related References:

For the STILT-ECMWF methodology
see e.g. Vardag et al., Biogeosciences (2016)

For the FLEXPART-COSMO methodology
see e.g., Henne et al., Atmos. Chem. Phys. (2016)

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Simulations key-points

Transport Simulations

2 Lagrangian particle dispersion models

- FLEXPART-COSMO (7×7 km), backward mode, 4 days
- STILT-ECMWF (10×10 km), backward mode, 10 days

2009-2017 (9 years)

3-hourly time-resolution

Boundary Conditions

Jena CarboScope CO_2 flux estimates
https://www.bgc-jena.mpg.de/CarboScope/

$\delta^{13}C$-CO_2 background relationship derived from atmospheric observations

CO_2 fluxes and $\delta^{13}C$ signatures

Anthropogenic Emissions: EDGARv4.3 "fuel"

base year 2010

scaled with annual BP fuel statistics and TNO time-factors

14 output categories:

- fossil: oil (3), coal (3), gas (2),
- biogenic: biofuels (1, liquid), biomass (1, solid), biogas (1)
- others: cement (1), chemical industry (1), metallurgy (1)

Biosphere Fluxes: Vegetation Photosynthesis & Respiration Model

2 output categories for 8 plant functional types:

- biospheric gross respiration («resp»)
- gross ecosystem exchange («gee»)

$\delta^{13}C$-CO_2 source signatures values attributed as noted in Table 1

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Model domains for FLEXPART-COSMO and STILT-ECMWF

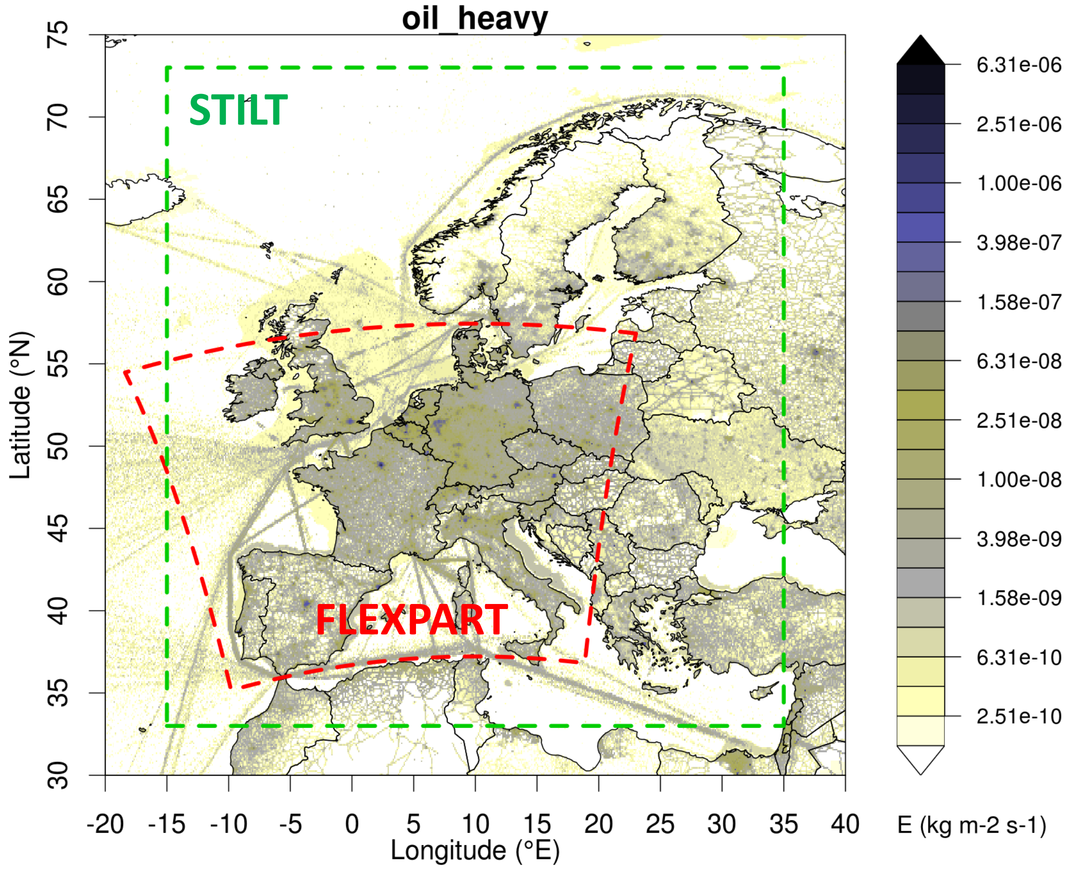

Figure 2. STILT-ECMWF (green) and FLEXPART-COSMO (red) model domain boundaries with underlying map of heavy oil related CO_2 emission fluxes from the anthropogenic emissions inventory (EDGAR v4.3 pre-release, fuel based). Emission fluxes correspond to base year 2010 here.

Related References:

For the STILT-ECMWF methodology
see e.g. Vardag et al., Biogeosciences (2016)

For the FLEXPART-COSMO methodology
see e.g., Henne et al., Atmos. Chem. Phys. (2016)

Anthropogenic Emissions Inventory:
EDGAR v4.3 pre-release, fuel-based, base year 2010

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$\delta^{13}C_s$ values used to simulate ambient $\delta^{13}C_a$

Table 1. Considered source signature values*, $\delta^{13}C_s$, per category.

	$\delta^{13}C_s$
FUEL USE: ***CO2.fuel***	
OIL	-26.5‰
OIL_heavy	
OIL_light	
OIL_mixed	
GAS	-44.0‰
GAS_natural	
GAS_derived	
COAL	-24.1‰
COAL_hard	
COAL_brown	
COAL_peat	
BIOGENIC FUELS	
BIOMASS (solid)	-24.1‰
BIOGAS (gas)	-60.0‰
BIOFUEL (liquid)	-26.5‰
OTHER SOURCES: ***CO2.cement***	
CEMENT PRODUCTION	-0‰
ECOSYSTEM PROCESSES: ***CO2.bsp*** (seasonally variable)	
BIOSPHERE RESPIRATION	-27 to -22‰
PHOTOSYNTHETIC UPTAKE	-25 to -20‰

**Reference Scale:* Vienna PeeDee Belemnite (VPDB)

Literature based emissions source signatures, $\delta^{13}C_s$ in ‰

Simulated regional CO_2 concentrations, f_r, in ppm

$$\delta^{13}C_{mix} = \frac{\sum_{n=1}^{i} |f_r,i| \times \delta^{13}C_s,i}{\sum_{n=1}^{i} (|f_r,i|)} \qquad (1)$$

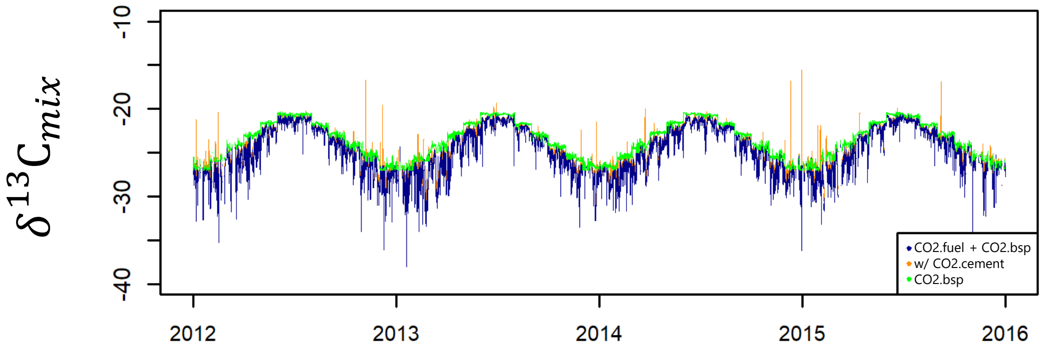

Figure 3. Simulated mixed $\delta^{13}C$-CO_2 emissions signatures on a 3-hourly time, here shown for the years 2012-2015. The mix of all categories (dark blue, i.e. *CO2.fuel* and *CO2.bsp*, and orange coloured trace, i.e. *CO2.cement*) following equation (1) is used in further in equation (2) when calculating simulated ambient $\delta^{13}C$-CO_2 ratios. The seasonality in the biospheric signatures (*CO2.bsp*, in green) yields an underying periodic pattern.

$$\delta^{13}C_a = \frac{(f_{bg} \times \delta^{13}C_{bg}) + (\sum_{n=1}^{i}(f_r, i) \times \delta^{13}C_{mix})}{f_{bg} + \sum_{n=1}^{i}(f_r, i)} \qquad (2)$$

Background values for CO_2 concentration, f_{bg}, in ppm, and $\delta^{13}C$-CO2 ratio, $\delta^{13}C_{bg}$, in ‰

Related References:
e.g.,
CDIAC at https://cdiac.ess-dive.lbl.gov/
Vardag et al., Biogeosciences (2016)

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Example: Short-Term Event $\delta^{13}C-CO_2$ Simulation

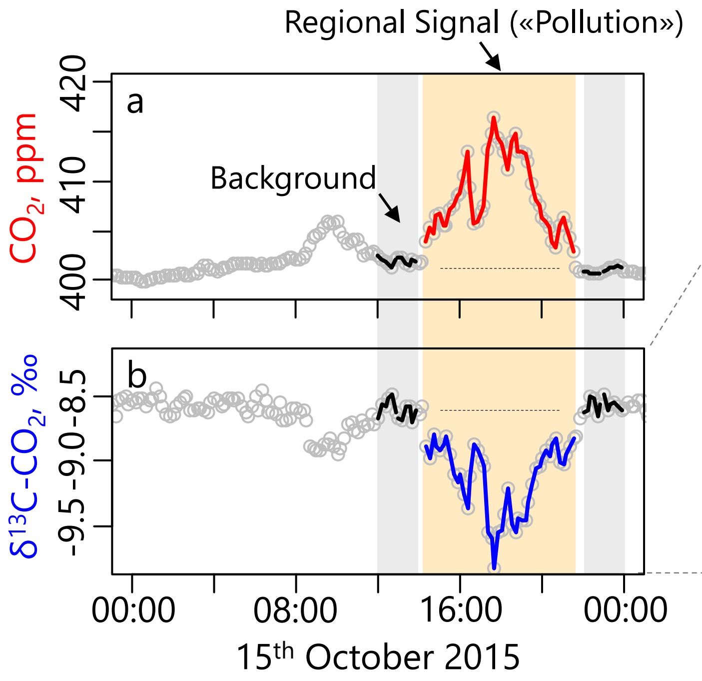

Figure 4. Representative time-series of continuous observations (10 minutes averages) of CO_2 concentration (a) and $\delta^{13}C-CO_2$ ratio (b) by in-situ QCLAS measurements.

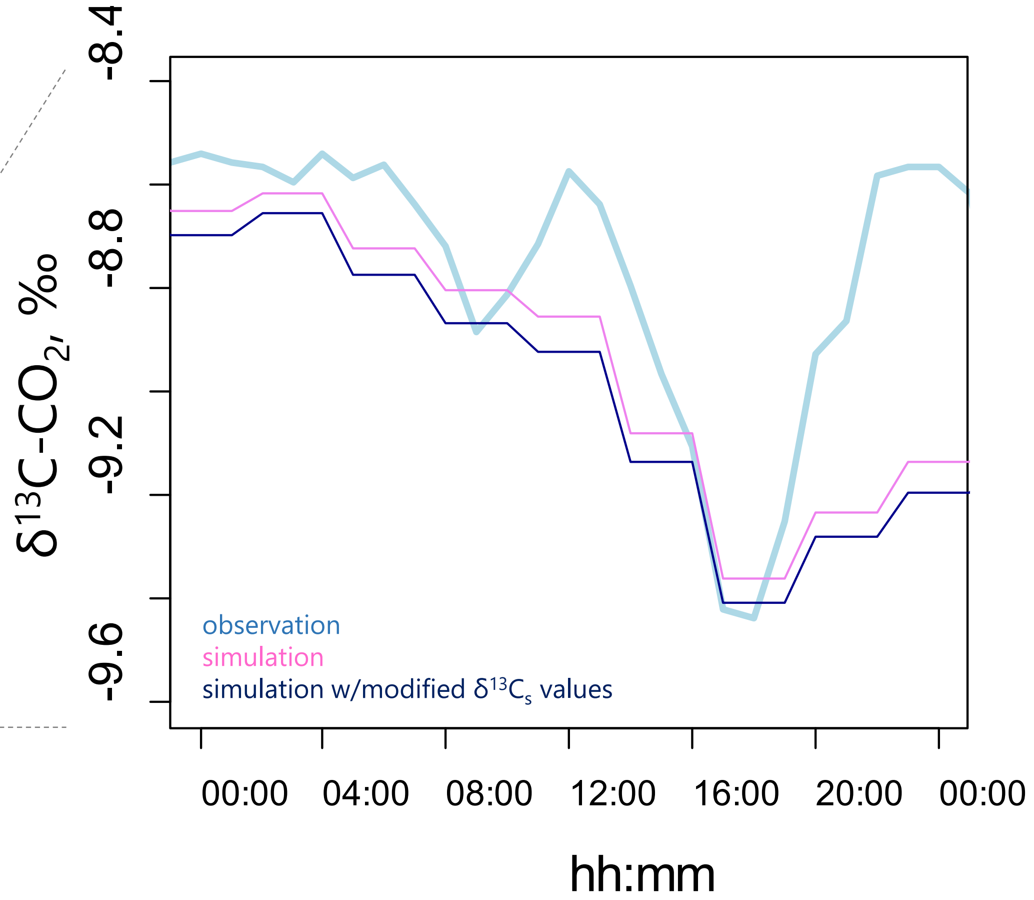

Figure 5. Simulations of $\delta^{13}C-CO_2$ in pink and dark blue colour following equation (2) in comparison to observations from Figure 4b on a 3-hourly time resolution (in light blue colour).

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Results

Simulation Performance

The simulated atmospheric CO_2 and $\delta^{13}C$-CO_2 time-series are in good agreement with the observations and capture the observed variability at the models' 3-hourly time-resolution, for both, the intensity of the regional signals, as well as their time-profile.

CO_2 contribution analysis

The simulations allow for an in-depth evaluation of the contribution of different CO_2 emission sources when Jungfraujoch is influenced by air masses from the planetary boundary layer (PBL):

- The receptor-oriented model simulations indicate that anthropogenic activities that influence the regional CO_2 concentrations at JFJ are primarily of fossil origin (90%).
- Anthropogenic activities typically make up 60% of the regional CO_2 concentrations simulated at JFJ in February.. They can be as low as 20% in July and August. The rest is attributed to gross biospheric CO_2 respiration.
- The gross biospheric CO_2 uptake outweighs regional CO_2 concentrations at JFJ from both, gross biosphere respiration, as well as anthropogenic activities. The effect is particularly large during June, July and August.

These findings are a peculiarity of the station's location in a remote Alpine area. They are, among other things, related to the fact that air masses lifted from the PBL frequently move through highly vegetated areas before reaching Jungfraujoch.

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Thank you!

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https://www.empa.ch/web/s503/research

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ICOS Carbon Portal

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Tuzson B, et al, 2008a: https://link.springer.com/article/10.1007/s00340-008-3085-4
Tuzson B, et al, 2008b: https://doi.org/10.1016/j.infrared.2007.05.006
Tuzson B, et al, 2011: https://doi.org/10.5194/acp-11-1685-2011
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$\delta^{13}C$-CO_2 source signatures
e.g.,
CDIAC, online at https://cdiac.ess-dive.lbl.gov/
Vardag SN, et al, 2015 : https://doi.org/10.5194/acp-15-12705-2015
Vardag SN, et al, 2016. : https://doi.org/10.5194/bg-13-4237-2016