

1 The authors like to thank the editor for his comments and suggestions. The Editor's comments
2 are given in bold, the authors' answers are formatted as plain text and changes to the
3 manuscript are given in italics.

4 **Editor Decision: Reconsider after minor revisions (Editor review) (22 Jun 2016) by**
5 **Andreas Engel**

6 **Comments to the Author:**

7 **The authors have addressed most issues raised by the reviewers. There are a few points**
8 **where I suggest some additional clarification:**

9 *** The new Figure 9 needs some explanation. Please explain more carefully what the**
10 **surface residence time is and how it is calculated.**

11 At page 8, line 23 we added the following

12 *"...source sensitivities, represented by total residence time, which were calculated as total*
13 *residence times derived by summing residence times over all start times and over all*
14 *integration time steps within a selected integration interval for all grid cells as given in*
15 *Henne et al. (2010)."*

16 **Reviewer #2 questioned the usefulness of figure 6-8. In general it seems to me that the**
17 **main message of these two figures is now condensed in Figure 9. I suggest to consider**
18 **either leaving out figures 6-8 or explaining them a little more. Right now they are**
19 **presented and very little discussion is actually given to the figures.**

20 Since figure 9 is not just a summary of figure 6-8 but gives additional information we decided
21 to keep all figures but changed and extended the discussion of figures 6-8 as follows:

22 *"Sensitivity analyses revealed that the upper tropospheric air originates from different*
23 *geographic regions, mainly from south west, than the in-situ air measured by the NDIR.*
24 *During summer, the NDIR measurements record mainly air from European regions, whereas*
25 *the FTIR sees more influence from the west (Figure 6). From winter to spring, NDIR CO₂*
26 *values are again driven by European sources, whereas FTIR values represent a significantly*
27 *wider foot print reaching to west and further to the north in contrast to the summer situation*
28 *(Figure 7, Figure 8). Similar studies investigating CO at JFJ also showed that JFJ is not only*
29 *sensitive to Central Europe but also to regions as far west as for example North America, the*
30 *Pacific or even Asia and that the influence of these regions is getting stronger with increasing*

1 height (Dils et al., 2011; Pfister et al., 2004; Zellweger et al., 2009). Therefore the air
2 measured by the FTIR is partially decoupled from the increasing CO₂ values of the winter-
3 time northern hemisphere. Furthermore, the decoupling might be amplified by the weak
4 overturn of tropospheric air in winter. Towards spring, the tropospheric overturn speeds up
5 again which results in synchronous CO₂ minima for both datasets in August (Figure 9). These
6 findings can help to understand the shift in the observed wintertime maximum of CO₂ between
7 FTIR (January) and NDIR (March-April). The land surfaces of northern hemispheric
8 midlatitudes act as a net CO₂ source during the winter half year, since photosynthesis is
9 largely reduced and respiration and anthropogenic emissions of CO₂ dominate the budget.
10 Hence, the maximum of CO₂ is observed at the end of the winter half year and close to the
11 surface. For the free troposphere above JFJ as observed by the FTIR the direct link to these
12 wintertime releases of CO₂ is weakened due to generally reduced vertical transport. At the
13 same time more frequent transport from and land surface contact in the tropics can be
14 deduced (Figure 9), an area that even during the winter half year may act as a net CO₂ sink
15 due to photosynthetic uptake.”

16 **The answer to the remark of reviewer #2 regarding the seasonality of the UTLS is not**
17 **satisfactory. I suggest to at least discuss the difference in seasonality between**
18 **stratosphere and troposphere and point out that this will influence the column but not**
19 **the ground measurements, providing a further possible shift in seasonality. I think that**
20 **the magnitude of the possible effect can be discussed without the need for a large model**
21 **study.**

22 We consulted several publications related to this topic namely Nakazawa et al. (1990), Hoor
23 et al., (2004), Ray et al. (2014), Engel et al. (2006), Sawa et al. (2008), Sawa et al. (2015).
24 These publications document a consistent anti-correlation between tropospheric and
25 stratospheric CO₂ mole fractions. There are two effects that were put forward to explain this
26 anti-correlation namely (i) the mixing of uplifted tropical air masses horizontally transported
27 to northern latitudes with locally uplifted tropospheric air; (ii) only vertical transport from the
28 troposphere through the tropopause into the lowermost stratosphere resulting in a damped and
29 time lagged stratospheric CO₂ signal.

30 From these publications we would guess that the influence of the tropical air transport to
31 latitudes of our investigations (46°N) has a minor influence on the phasing between
32 tropospheric and stratospheric CO₂ evolutions, due to the small phase shift of about 2 months

1 in the CO₂ signal between the tropics and the mid-latitudes. Therefore we expect the direct
2 vertical transport through the tropopause to be the main influence for the observed phase shift
3 between the tropospheric and stratospheric CO₂. Based on that, we estimate that the FTIR
4 measurements represent a pressure weighted mean of tropospheric and stratospheric CO₂
5 mole fractions being anti-correlated. A simple calculation assuming the stratospheric signal to
6 be anti-correlated and damped to an amplitude of 30% (4.1 ppm) compared to the in-situ
7 observations at Jungfraujoch leads to an amplitude that corresponds rather nicely with the
8 FTIR observations, which is 4.4 ppm. The evolution of CO₂ mole fraction over the course of a
9 year results in an excellent agreement with the FTIR observations during summer, which
10 seems to be mainly influenced by direct vertical transport. In contrast, during spring there is a
11 substantial mismatch of about 1.5 – 2 ppm.

12 Since these are very simple calculations we hesitate to include them into the manuscript.
13 However, we would agree to change the manuscript accordingly if you as editor ask for it. In
14 this case we would suggest a similar text to be included as shown above.

15 **The change suggested by the author on p. 15. l 27 regarding the non-exploitation of the**
16 **vertical resolution of the FTIR data is misleading, as it may suggest that the FTIR has a**
17 **good vertical resolution which was just not exploited. Please merge the old and the new**
18 **statement, explaining that vertical resolution is poor and was therefore not exploited.**

19 The sentence was changed from:

20 *“...or (c) since the FTIR vertical sensitivity was not exploited in the present retrievals the*
21 *measured column signal contains mixed information from the troposphere and the*
22 *stratosphere.”*

23 to:

24 *“...or (c) since the FTIR has a low vertical sensitivity it was not exploited in the present*
25 *retrievals and therefore the measured column signal contains mixed information from the*
26 *troposphere and the stratosphere.”*

27

28 Change on the authors' behalf:

29 Page 10, line 11 was changed from:

30 *“... the FTIR columns and were especially increased...”*

1 To:

2 “...the FTIR columns compared to the surface and were especially increased...”

3

4 References:

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28

1 **Intercomparison of in-situ NDIR and column FTIR**
2 **measurements of CO₂ at Jungfrauoch**

3
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12 Keywords: CO₂, FTIR, NDIR, Jungfrauoch, intercomparison, CO₂ trend

13 **Abstract**

14 We compare two CO₂ time series measured at the High Alpine Research Station Jungfrauoch
15 (3580 m a.s.l., Switzerland) in the period from 2005 to 2013 with an in-situ surface
16 measurement system using a nondispersive infrared analyzer (NDIR) and a ground-based
17 remote sensing system using solar absorption Fourier Transform Infrared spectrometry
18 (FTIR). Although the two data sets show an absolute shift of about 13 ppm, the slopes of the
19 annual CO₂ increase are in good agreement within their uncertainties. They are 2.04 ± 0.07
20 ppm yr^{-1} and $1.97 \pm 0.05 \text{ ppm yr}^{-1}$ for the FTIR and the NDIR system, respectively. The
21 seasonality of the FTIR and the NDIR system is $4.46 \pm 1.11 \text{ ppm}$ and $10.10 \pm 0.73 \text{ ppm}$,
22 respectively. The difference is caused by a dampening of the CO₂ signal with increasing
23 altitude due to mixing processes. While the minima of both data series occur in the middle of
24 August, the maxima of the two datasets differ by about ten weeks, the maximum of the FTIR
25 measurements is in middle of January, whereas the maximum of the NDIR measurements is
26 found at the end of March. Sensitivity analyses revealed that the air masses measured by the
27 NDIR system at the surface of Jungfrauoch are mainly influenced by central Europe, whereas
28 the air masses measured by the FTIR system in the column above Jungfrauoch are influenced
29 by regions as far west as the Caribbean and the United States.

1 The correlation between the hourly averaged CO₂ values of the NDIR system and the
2 individual FTIR CO₂ measurements is 0.820, which is very encouraging given the largely
3 different sampling volumes. Further correlation analyses showed, that the correlation is
4 mainly driven by the annual CO₂ increase and to a lesser degree by the seasonality. Both
5 systems are suitable to monitor the long-term CO₂ increase, because this signal is represented
6 in the whole atmosphere due to mixing.

7

8 **1 Introduction**

9 CO₂ is the most important anthropogenic greenhouse gas, with a large contribution to the
10 greenhouse effect (Arrhenius, 1896) and an additional radiative forcing of the atmosphere
11 currently evaluated at 1.68 Wm⁻² (IPCC, 2013). The strength of the forcing is depending on
12 its atmospheric mole fraction which is ruled by the processes of the carbon cycle as well as by
13 anthropogenic CO₂ emissions from fossil fuel combustion and land use change. The major
14 reservoirs of the carbon cycle besides the lithosphere are the soils, the ocean, the biosphere
15 and the atmosphere, where the latter is also acting as the main link between the biosphere and
16 the ocean. The linking process between the atmosphere and the ocean is dissolution of CO₂ in
17 oceanic water, where it is subsequently chemically bound to bicarbonate and carbonate and
18 therefore removed from the carbon cycle on a longer timescale (Broecker and Peng,
19 1982;Feely et al., 2004;Heinze et al., 1991;Sillén, 1966). The processes coupling the
20 biosphere with the atmosphere are photosynthesis, where CO₂ is taken up by plants, and
21 respiration, where CO₂ is released back to the atmosphere. Photosynthesis and respiration are
22 mainly driven by climatic conditions of the environment. In the northern hemisphere,
23 especially in the extratropics with distinct seasons, the dominating process in late spring,
24 summer and fall is photosynthesis and thereby the uptake of CO₂ from the atmosphere. In
25 autumn respiration and with it the release of CO₂ from the biosphere into the atmosphere
26 starts to take over and is the ruling process in winter until spring when photosynthesis
27 becomes the dominating process again. Due to these alternating processes, the CO₂ mole
28 fraction in the atmosphere shows a seasonal cycle with its maximum generally in early spring
29 and its minimum in fall (Halloran, 2012;Keeling et al., 1976;Keeling et al., 2001;Machida et
30 al., 2002). A further component in the change of atmospheric CO₂ mole fraction is CO₂
31 release due to fossil fuel combustion (Karl and Trenberth, 2003;Revelle and Suess, 1957;Tans
32 et al., 1990). Nowadays, roughly half of the anthropogenically produced CO₂ ends up in the

1 oceans and the biosphere, whereas the other half is accumulating in the atmosphere and leads
2 to a more or less steady increase of the atmospheric CO₂ mole fraction (Bender et al., 2005;Le
3 Quéré et al., 2013;Sabine et al., 2004). Measuring the atmosphere's CO₂ mole fraction on the
4 long-term is therefore important to understand the sources and sinks of the carbon cycle and
5 the annual CO₂ increase due to fossil fuel combustion and land use change. To measure the
6 evolution of CO₂ in the atmosphere on a global scale satellite remote sensing methods can be
7 used as e.g. OCO-2 (Crisp et al., 2004, Pollock et al., 2010, Thompson et al., 2012) or
8 GOSAT (Chevallier et al., 2009, Yokota et al., 2009) but they are limited by e.g. cloud cover,
9 temporal coverage due to the orbit, coarse resolution etc. An intercomparison between
10 GOSAT and several TCCON (Total Carbon Column Observation Network) stations showed a
11 mean difference for daily averages of -0.34 ± 1.37 ppm (Heymann et al., 2015). Ground based
12 measurement systems on the other hand have a high temporal resolution and provide very
13 accurate data, which can be used to validate satellite data (Buchwitz et al., 2006; Butz et al.,
14 2011;Dils et al., 2006; Morino et al., 2011;Wunch et al., 2011) or as model input (Chevallier
15 et al., 2010), but surface observations have often a limited representativeness and are often
16 influenced by nearby processes and hence, not representative for larger areas. Also the
17 influence of the biosphere or anthropogenic pollution can be a serious issue and make it very
18 challenging to measure background air. Therefore, to measure global CO₂ trends the sampling
19 site should be at a very remote place like e.g. Mace Head Station (Bousquet et al.,
20 1996;Messenger et al., 2008) on the western coast of Ireland or the flask sampling network in
21 the Pacific of NOAA (Komhyr et al., 1985;Troler et al., 1996). Another possibility is to
22 measure in the free troposphere e.g. with airplanes as done in the CARIBIC project
23 (Brenninkmeijer et al., 2007) or the CONTRAIL project (Machida et al., 2008) or at high
24 altitudes which are mostly in the free troposphere as e.g. Mauna Loa (Keeling et al.,
25 1976;Keeling et al., 1995;Pales and Keeling, 1965;Thoning et al., 1989). The High Alpine
26 Research Station Jungfraujoch (JFJ) with its altitude of 3580 m a.s.l. (Sphinx Observatory)
27 and position mostly above the planetary boundary (Henne et al., 2010) is therefore a very
28 suitable spot to conduct ground based CO₂ background measurements.

29 The University of Liège (Belgium) has been measuring infrared radiation at JFJ since the
30 1950s and started regular FTIR (Fourier Transform InfraRed) measurements in 1984. The
31 Climate and Environmental Physics Division (KUP) of the University of Bern started
32 measuring CO₂ and δO₂/N₂ in 2000 by a flask sampling program and since the end of 2004,
33 CO₂ and O₂ have been additionally measured with a continuously operating system of a NDIR

1 instrument and a paramagnetic cell. In this study we compared the FTIR and the NDIR data
2 set to see if the two complementary measurement techniques are catching the same trends,
3 seasonalities and variations in atmospheric CO₂ mole fraction at and above Jungfraujoch.

4

5 **2 Methods**

6 **2.1 Measurement site**

7 The High Altitude Research Station Jungfraujoch (JFJ) is located 7°59'02'' E, 46°32'53'' N
8 at the northern margin of the Swiss Alps. The Jungfraujoch is a mountain saddle between the
9 Mönch (4099 m a.s.l.) and Jungfrau (4158 m a.s.l.) summits at a height of 3580 m a.s.l.
10 (Sphinx Observatory) and is accessible year-round by train. Because of the high elevation, the
11 station is usually above the planetary boundary layer (PBL) and therefore mainly receives air
12 from the free troposphere which is why it was classified as “mostly remote” by Henne et al.
13 (2010). Nevertheless, the station can be influenced by polluted air during specific events such
14 as frontal passages and Föhn (Uglietti et al., 2011; Zellweger et al., 2003) or thermal uplift of
15 polluted air from the surrounding valleys on fair weather days (Baltensperger et al., 1997;
16 Henne et al., 2005; Zellweger et al., 2000). Because of the high elevation, the accessibility and
17 the good infrastructure, the JFJ is an ideal location for in-situ measurements of atmospheric
18 background air from continental Europe (Baltensperger et al., 1997; Henne et al.,
19 2010; Zellweger et al., 2003). JFJ is also one of the currently 29 core sites of the WMO GAW
20 (Global Atmospheric Watch) programme.

21 **2.2 In-situ NDIR measurements at Jungfraujoch**

22 The KUP CO₂ measurements are based on a combined system to monitor CO₂ and O₂
23 changes in the atmosphere. The ambient air is entering through a strongly ventilated (600
24 m³ h⁻¹) common inlet on the observatory's roof to a manifold, which serves many trace gas
25 analyzers, where an aliquot of it is drawn to the KUP system. The air is cryogenically dried to
26 a dew point of -90 °C (FC-100D21, FTS systems, USA). Temperature as well as pressure is
27 stabilized to avoid influences caused by ambient air density fluctuations. This allows the
28 determination of CO₂ by a NDIR spectrometer (Maihak S710) measuring at a wavelength of
29 4.26 μm with a frequency of 1 Hz and O₂ by a paramagnetic cell under highly controlled
30 conditions. Measurements are done in a cyclic sequence of 18 hours with each gas measured

1 for 6 minutes with only the last 115 seconds of a six minute period used for mole fraction
2 determination, to allow for signal stabilization after changing the sample source. At the
3 beginning of each 18-hour sequence, the system is calibrated with two reference gases (high
4 and low span). A working gas is measured between two ambient air measurements to correct
5 for short term variations. All measurements ending in a particular hour are used for the
6 calculation of hourly mean CO₂ observations, which in our case includes therefore 6 ambient
7 observation values per hour. Cylinder measurements with a known mole fraction showed a
8 long-term precision for hourly averages better than 0.04 ppm. The accuracy of our target
9 cylinder corresponds to less than 0.1 ppm (WMO target value for CO₂ measurements)
10 calculated as standard deviation of the mean considering the number of independent
11 calibration set (high span, low span, working gas). The CO₂ values are reported on the WMO
12 X2007 scale. A multi-annual intercomparison between the NDIR system and a cavity ring-
13 down spectroscopy at JFJ showed a very good agreement of the CO₂ measurements (Schibig
14 et al., 2015).

15 **2.3 Column FTIR measurements at Jungfraujoch**

16 The University of Liège has been recording atmospheric solar spectra at JFJ since the early
17 1950s. The current FTIR instrument is a commercially available Bruker IFS-120 HR with a
18 resolution of up to 0.001 cm⁻¹ (Mahieu et al., 1997). It features interchangeable detectors, a
19 KBr beam-splitter and dedicated optical filters, which altogether give the possibility to cover
20 the 1 to 14 μm spectral range (Zander et al., 2008). Here gases such as CO₂, CH₄ and H₂O
21 show numerous absorption lines documenting contributions to the greenhouse effect. These
22 spectra also contain information about the abundance of many additional absorbing gas
23 species in the path between the instrument and the sun, essentially present either in the
24 troposphere or in the stratosphere. The CO₂ data set used here has been derived from the
25 reference total column time series produced within the framework of the NDACC monitoring
26 program (Network for the Detection of Atmospheric Composition Change; see
27 <http://www.ndacc.org>), presented previously in e.g. Zander et al. (2008; see Figure 6). In the
28 meantime, the data set has been consistently updated, still using the SFIT-1 algorithm
29 (version 1.09c) and a single microwindow spanning the 2024.3 – 2024.7 cm⁻¹ spectral
30 interval, whose main spectral line at 2024.564 cm⁻¹ is coming from ¹³CO₂. The uncertainty
31 range on the strength of this CO₂ line is estimated at 2 to less than 5 % in the HITRAN
32 compilation (Rothman et al., 2005), leading to a systematic error on the retrieved total column

1 of the same magnitude. The single CO₂ a priori vertical distribution used in all retrievals is
2 characterized by a constant mixing ratio of 338 ppm from the surface up to the tropopause,
3 then slightly decreasing to stabilize at 330 ppm at 20 km and above. During the retrieval
4 process, a simple scaling of the whole vertical profile is performed, accounting for
5 interferences by weak ozone and water vapor lines, and the mixing ratio derived for CO₂ in
6 the troposphere is used in the present comparisons. Note that the representativeness of this
7 unique profile is not optimal for all seasons and may lead to an underestimation of the
8 seasonal amplitude (see Fig. 1 in Barthlott et al., 2015), because of a non-optimum vertical
9 sensitivity of the FTIR retrieval. Indeed, typical values of the total column averaging kernel –
10 indicative of the fraction of information coming from retrieval rather than from the a priori
11 (e.g. Vigouroux et al., 2015) – are in the 0.5 – 1 range between the ground and 10 km altitude,
12 in line with Fig. 4 of Barthlott et al. (2015). Over all the standard deviation of multiple
13 measurements over the course of a single day corresponds to less than one ppm, which is
14 significantly smaller than the observed seasonal cycle.

15 **2.4 Data processing**

16 The NDIR data set is much more influenced by near ground processes like thermal uplift of
17 PBL air from the surrounding valleys, advection of PBL air by synoptic events etc. than the
18 FTIR and shows therefore a higher variability. Additionally, because of the large volume of
19 the column sampled by the FTIR above JFJ the CO₂ mole fraction measured by the FTIR is
20 averaged and the data set is far less sensitive to local events than the in-situ NDIR
21 measurements. The FTIR needs a cloudless sky to be able to measure, whereas the NDIR
22 system is measuring under all conditions, which can lead to very high CO₂ mole fractions
23 during e.g. Föhn events, when the sky is cloudy and polluted air from the heavily
24 industrialized Po basin (Northern Italy) is advected to JFJ. Therefore, only measurements of
25 background air should be taken into account to compare the two data sets properly.

26 **2.4.1 Filtering, trend and seasonality calculation**

27 The background data were selected using a statistical approach. A cubic spline was fitted to
28 both datasets individually, the standard deviation of the residuals was calculated and all points
29 beyond 2.7 σ were flagged as outliers. This process was repeated in both data sets until
30 convergence. The threshold of 2.7 σ was chosen because in normally distributed data more

1 than 99 % of the total data points would be included for further calculations and only the most
2 obvious outliers (less than 1 %) would be rejected.

3 The CO₂ mole fraction is dominated by two major processes. One is the linear increase due to
4 fossil fuel combustion (trend) and one is the annual in- and decrease due to respiration and
5 photosynthesis, and to a lesser degree due to fossil fuel combustion (seasonality). The trend
6 was calculated for both datasets individually with a Monte Carlo approach.

7 For the trend calculation we intentionally used the datasets including seasonal signals because
8 it leads to realistic trend error estimates compared to deseasonalized datasets, which in our
9 view tend to underestimate the error. The datasets were split in two subsets, where each of the
10 subsets spanned over $n - 0.5$ phases (in this study n equals 9 years) to prevent a bias in the
11 trend calculation due to the seasonal cycle. The first subsets start in January 2005, the second
12 subsets start in July 2005. In each subset about 2 % (a higher number does improve the result)
13 of the points were selected randomly and the linear trend was calculated. This was repeated
14 500 times with each subset and the averages of these linear trends were taken as the slopes of
15 the datasets.

16 To calculate the seasonality, the two datasets were detrended and monthly averages were
17 formed, from which the seasonality was calculated as the difference between the highest and
18 the lowest value.

19 **2.4.2 Correlation analysis**

20 Because of the different time resolutions for in-situ and FTIR measurements we selected
21 those in-situ measurements (six minute and hourly NDIR averages) that are closest (± 30 min)
22 to the FTIR values for correlation analysis.

23 Since the differences between both correlation analyses were negligible (see results section),
24 it was decided to continue with the hourly averages of the NDIR dataset only, which is the
25 common output of the NDIR database.

26 The FTIR's sample volume is much bigger than the NDIR system's and because of
27 transportation processes there's a possibility of mixing processes. To check, a moving average
28 of the NDIR data with increasing width was calculated to see if the correlation is enhanced
29 with expanding width (from 0 to ± 600 h).

30 Furthermore, the column measurements were retrieved for the layer between 3.58 km (altitude
31 of the Sphinx Observatory) to the top atmosphere (set to 100 km in the retrieval scheme)
32 whereas the NDIR system is measuring at the lower boundary of the FTIR's sampling

1 column, therefore it is possible that a time shift in the measured CO₂ mole fractions due to
2 advection, uplift of air parcels etc. occurs. To check whether a systematic time shift exists
3 between the two datasets, the NDIR measurements were shifted relative to the FTIR data
4 from -60 to +60 days (corresponding to -1440 h to +1440 h) in hourly steps and again the
5 correlation of the two data sets was calculated. If there is a systematic time shift, the deviation
6 should be indicated by increased correlation values.

7 **2.5 FLEXPART model runs**

8 From 2009 to 2011, backward Lagrangian particle dispersion model simulations were
9 performed with FLEXPART (Stohl, et al. 2005) to simulate the transport towards JFJ and
10 estimate surface source sensitivities (footprints) of the sampled air masses. To account for the
11 complex flow in the Alpine area, a regional scale version of the model driven by operational
12 output from the regional scale numerical weather prediction model COSMO as produced by
13 MeteoSwiss was used (Henne et al., 2015, Oney et al., 2015) . Since COSMO is a limited area
14 model, the transport of particles leaving the domain was further simulated in the global scale
15 version of FLEXPART (Stohl et al., 2005) driven by operational analysis fields of the
16 European Centre for Medium Range Weather Forecast (ECMWF). In the Alpine area,
17 COSMO input data had a horizontal resolution of approximately 2 km x 2 km, in Western
18 Europe 7 km x 7 km. Of the 1214 FTIR measurements in this period, footprints were
19 available for 766. The model simulated footprints of the surface in-situ observations and five
20 partial columns above JFJ reaching from 3365-4226 m a.s.l., 4226-4912 m a.s.l., 4912-5629
21 m a.s.l., 5629-6386 m a.s.l. and 6386-7184 m a.s.l. The lower boundary is below JFJ in order
22 to account for smoothed model topography. Particles released at and above JFJ were followed
23 10 days backward in time to calculate source sensitivities, represented by total residence time,
24 which were calculated as total residence times derived by summing residence times over all
25 start times and over all integration time steps within a selected integration interval for all grid
26 cells as given in Henne et al. (2010). Source sensitivities were evaluated on regular
27 longitude/latitude grids. The resolution was 0.5° x 0.5° globally, 0.2° x 0.2° over Europe and
28 an even higher resolution of 0.1° x 0.1° was used in the Alpine area. The footprints of the
29 individual measurements of each partial column were averaged to monthly means to get
30 information about the origin of the air masses in the according month (Henne, 2014;Henne et
31 al., 2013).

32

1 3 Results

2 Because of the different measurement techniques, the number of data points in the two
3 datasets is different. In the period 2005 to 2013 the NDIR dataset contains 68477 hourly
4 averages from which about 5 % were omitted as pollution or depletion events resulting from
5 PBL influence as estimated by the filtering (Figure 1). In the same period, the FTIR dataset
6 shows 3068 measurements of which about 5 % were rejected as pollution and depletion
7 events, too (Figure 2). For all further calculations, only the filtered datasets were used.

8 The average of the detrended and deseasonalized NDIR data before and after filtering was
9 0.00 ± 2.65 ppm and 0.00 ± 1.84 ppm (Figure 3 A), the average of the FTIR data was $0.01 \pm$
10 2.61 ppm and 0.01 ± 2.16 ppm, respectively (Figure 3 B).

11 With a Monte Carlo algorithm, the values of the annual change of the CO₂ mole fraction of
12 the two datasets were calculated. Despite the shift between the two datasets of roughly 13
13 ppm and the different measurement techniques the annual CO₂ increase is quite similar. The
14 FTIR slope is 2.04 ± 0.07 ppm yr⁻¹ and the NDIR dataset shows a slope of 1.97 ± 0.05 ppm
15 yr⁻¹, so they are equal within their uncertainties (Figure 4). The observed offset between the
16 FTIR (NDACC) and in-situ records at Jungfraujoch contrasts the comparison of NDACC and
17 TCCON records as determined at Ny-Ålesund which do not show any offset at all when using
18 several individual CO₂ lines for the mid-IR (Buschmann et al., 2016). However, the
19 FTIR/NDIR offset of about 3% is commensurate with the systematic uncertainty affecting
20 the FTIR measurement; see section 2.3.

21 By detrending the datasets with the derived slopes, the seasonality can be calculated. The
22 column dataset shows a seasonality of 4.46 ± 1.11 ppm whereas the in-situ measurements at
23 the Sphinx Observatory show a seasonality roughly twice as big, namely 10.10 ± 0.73 ppm. To
24 find the moment of the average minima and maxima, a two harmonic fit function was applied
25 to the detrended datasets. The minima of the FTIR and NDIR datasets are both in the middle
26 of August, but the maxima are roughly ten weeks apart. The maximum of the NDIR datasets
27 occurs at the end of March, whereas seasonality of the FTIR dataset already reaches its
28 maximum in the middle of January (Figure 5).

29 The footprints of August, January and March, when the extrema of the seasonal cycle
30 occurred, as calculated with FLEXPART show that the in-situ observation at Jungfraujoch is
31 mainly receiving air masses that are influenced by Central Europe, and to a lesser degree by
32 the Mediterranean area and the northern Atlantic (Figure 6, Figure 7 and Figure 8).

1 With increasing altitude, the footprints of the sub-columns indicate, that the measured air
2 masses become more sensitive to regions as far west as e.g. the Caribbean and the United
3 States and that the influence from the European continent and northern regions higher than
4 50°N is decreasing (Figure 6, Figure 7 and Figure 8).

5 In general, the decoupling between the FTIR columns and possible surface fluxes of CO₂
6 from land surfaces north of 30°N was strongest during the winter month (January to March),
7 when especially low surface residence times were simulated by FLEXPART for the free
8 tropospheric FTIR columns (Figure 9). From April to September larger surface residence
9 times were seen also for the FTIR columns and a stronger coupling between surface fluxes
10 and the free troposphere can be expected. At the same time residence times over tropical land
11 surface (south of 30°N) were generally larger for the FTIR columns compared to the surface
12 and were especially increased from February to April (see Figure 9).

13 To estimate the relationship between the FTIR and NDIR measurements the correlation was
14 calculated. The FTIR measurements take normally about 10 min and are done whenever
15 possible. Therefore the FTIR data is reported exactly at the measuring time. The NDIR on the
16 other hand is measuring non-stop, but only 115 s of six-minute intervals (see methods) are
17 used to calculate a data point and the six-minute data is normally averaged to hourly averages.
18 Therefore we first checked whether the high resolution data are necessary or hourly data is
19 good enough. To do so, to each FTIR data point the nearest high resolution and hourly
20 averaged NDIR values were assigned. An additional condition was that the NDIR value must
21 not be further apart than ± 30 min, otherwise no NDIR data point was set, which was the case
22 in about 10 % of the FTIR data points. The correlation between the FTIR and the high
23 resolution NDIR CO₂ measurements and between the FTIR and the hourly averages were
24 calculated to be 0.819 and 0.820, so the differences between the two regression values are
25 negligible. To examine the relationship between the FTIR and the NDIR measurements
26 further, the seasonality of the two datasets was eliminated which gave almost the same
27 correlation of 0.824 (0.838 with the high resolution data). In the next step only the trend was
28 subtracted and the remaining seasonalities were compared, which lead to a much smaller
29 correlation of 0.460 (0.461 with the high resolution data). In a final step, the trend as well as
30 the seasonality was removed, which resulted in a correlation of 0.071 (0.084 high resolution
31 data vs. FTIR). Since correlations between the FTIR data and the NDIR's high resolution and
32 the hourly data were almost the same, only the hourly data was considered for further
33 calculations (Figure 10).

1 As mentioned above, the column measurements represent the whole vertical distribution
2 above Jungfraujoch whereas the NDIR system is measuring at the base of the FTIR's
3 sampling column. Therefore, the two records might be time-delayed due to advection, uplift
4 of air parcels etc. To check for a potential time lag, the NDIR measurements were shifted
5 relative to the FTIR data from -1440 to +1440 hours in hourly steps.

6 The correlations between the NDIR and FTIR datasets and between the deseasonalized NDIR
7 and FTIR datasets show a peak region at a time shift from -10 h to 60 h with the highest
8 correlation being 0.830 and 0.836 respectively (Figure 11 A, Figure 11 B). The correlation
9 between the datasets is decreasing before and after this range, in the deseasonalized datasets
10 the correlation stays more or less stable. The correlation between the two trend corrected
11 datasets shows a plateau of enhanced correlation values from -50 h to 200 h time shift with a
12 maximum correlation of 0.495 at a time shift of 165 h, at lower and higher time shifts, the
13 correlation is decreasing (Figure 11 C). The correlation of the detrended and deseasonalized
14 datasets shows no distinct pattern and is oscillating around 0 (Figure 11 D).

15 Since the air volume measured by the FTIR is much bigger than the NDIR system's volume,
16 vertical mixing and transport processes can occur and thereby changing the CO₂ mole fraction
17 in the measured air parcels. Therefore moving averages with increasing widths (up to ± 600 h)
18 were calculated from the NDIR data and the obtained averaged NDIR values were correlated
19 with the filtered FTIR dataset. Changing the width of the moving average doesn't have a
20 strong influence on the correlation between the two filtered datasets, because the increasing
21 width of the moving average just smooths the dataset. The correlation remains at about 0.85
22 (Figure 12 A), with a very small increase of the correlation at the beginning, most probably
23 due to the above mentioned smoothing effect. The same is true for the correlation between the
24 deseasonalized datasets. They show high correlation of about 0.84 over the whole range of
25 widths, with a slight increase at the beginning, which is not significant (Figure 12 B). By
26 detrending the datasets, the correlation is increasing with the width of the moving average and
27 shows a plateau of higher correlation of about 0.5 at a width 150 to 600 h from where on it is
28 decreasing again (Figure 12 C). However, the changes in the correlation within the range of
29 150 h to 600 h are very small. The detrended and deseasonalized datasets show a very low
30 correlation and the improvement of the correlation due to the changing width of the moving
31 average is negligible. Over all, the improvement of the correlations due to the changing width
32 of the moving average is very small (Figure 12 D).

1 Finally both, the time shift and the width of the moving average were varied about ± 1440 h
2 and ± 600 h, to see with which combination of time shift and width the best correlation can be
3 reached. They all show a ridge of higher correlation at a time shift around zero which is
4 broadening with increasing width of the moving average, except for the data without slope
5 and seasonality, which have a low correlation anyway (Figure 13). The increasing width of
6 the moving average leads to a small improvement of the correlations in the beginning,
7 however over all it doesn't seem to have a strong influence on the correlations. The time shift
8 on the other hand has an influence on correlation between the complete filtered datasets and
9 even more on the correlation of the detrended datasets. In the correlation of the
10 deseasonalized datasets, the influence of the time shift is very limited except for the small
11 ridge of slightly enhanced correlations around zero time shift as mentioned above.

12

13 **4 Discussion**

14 The filtered FTIR and NDIR datasets show a very similar increase in the CO₂ mole fraction of
15 ambient air, despite the two totally different measurement principles. The calculated annual
16 CO₂ trends of the FTIR and NDIR datasets are 2.04 ± 0.07 ppm yr⁻¹ and 1.97 ± 0.05 ppm yr⁻¹
17 respectively (Figure 4) and are in good agreement with flask measurements done at JFJ with a
18 slope of 1.85 ppm yr⁻¹ (van der Laan-Luijkx et al., 2013) and other remote stations in the
19 northern hemisphere; for example Mauna Loa with 2.05 ppm yr⁻¹ (Tans and Keeling, 2014) or
20 Alert with 1.85 ppm yr⁻¹ (Keeling et al., 2001). Also the NDIR dataset's average seasonality
21 of 10.10 ± 0.73 ppm is in good agreement with the seasonality of these flask measurements,
22 which were 10.54 ± 0.18 ppm in the period 2007 to 2011 (van der Laan-Luijkx et al., 2013)
23 and is roughly double the FTIR's average seasonality of 4.46 ± 1.11 ppm (Figure 5). The
24 lower seasonality of the FTIR dataset can be explained by the fact that the NDIR system is
25 measuring CO₂ mole fractions at the Sphinx Observatory, which is most of the time above the
26 PBL (Henne et al., 2010) but still closer to the ground than the FTIR measurements.
27 Therefore the signal of the biosphere is stronger than in the column, where it is attenuated by
28 vertical mixing and transport processes of the atmosphere with increasing height. Also the
29 fixed a priori vertical CO₂ profile may contribute partly to the lower seasonality of the FTIR
30 measurements. The shape of the profile used to retrieve the CO₂ data doesn't reproduce the
31 changes due to seasonality and is therefore not always the optimum. By using a seasonally
32 varying a priori retrieval the seasonality might be slightly higher because the amplitude of

1 CO₂ is better retrieved (Barthlott et al., 2015). Furthermore, in the tropopause and the lower
2 stratosphere, the phase of the CO₂ seasonality is shifted by several months (Bönisch et al.,
3 2008; Gurk et al., 2008; Bönisch et al., 2009). However, this has only a minor influence on the
4 observed dampening of the amplitude of the FTIR seasonality compared to the vertical
5 mixing, since the stratosphere contains only about 10 % of the abundance of atmospheric air
6 molecules.

7 It is not easy to define the seasonal minimum and maximum in the FTIR dataset because they
8 are not very clearly pronounced. By fitting a two harmonic function the minimum was found
9 to be in the middle of August, the maximum in the middle of January. While the minimum of
10 the NDIR dataset is around the same time, the maximum of the FTIR dataset occurs roughly
11 ten weeks earlier than the maxima of the NDIR dataset (Figure 5). The timing of the minima
12 of both datasets and the maximum of the NDIR dataset coincide quite well with net land-
13 atmosphere carbon flux changes from negative to positive values and vice versa (Zeng et al.,
14 2014). Therefore an alternative explanation is needed for the early maximum of the FTIR
15 dataset. Sensitivity analyses revealed that the upper tropospheric air originates from different
16 geographic regions, mainly from south west, lower latitudes than the in-situ air measured by
17 the NDIR. During summer, the NDIR measurements record mainly air from European
18 regions, whereas the FTIR sees more influence from the west (Figure 6). From winter to
19 spring, NDIR CO₂ values are again driven by European sources, whereas FTIR values
20 represent a significantly wider foot print reaching to west and further to the north in contrast
21 to the summer situation (Figure 7, Figure 8). ~~Therefore the air measured by the FTIR is~~
22 ~~partially decoupled from the increasing CO₂ values of the winter time northern hemisphere.~~
23 ~~Furthermore, the decoupling might be amplified by the weak overturn of tropospheric air in~~
24 ~~winter. Towards spring, the tropospheric overturn speeds up again which results in~~
25 ~~synchronous CO₂ minima for both datasets in August.~~ Similar studies investigating CO at JFJ
26 also showed that JFJ is not only sensitive to Central Europe but also to regions as far west as
27 for example North America, the Pacific or even Asia and that the influence of these regions is
28 getting stronger with increasing height (Dils et al., 2011; Pfister et al., 2004; Zellweger et al.,
29 2009). Therefore the air measured by the FTIR is partially decoupled from the increasing CO₂
30 values of the winter-time northern hemisphere. Furthermore, the decoupling might be
31 amplified by the weak overturn of tropospheric air in winter. Towards spring, the tropospheric
32 overturn speeds up again which results in synchronous CO₂ minima for both datasets in
33 August (Figure 9). ~~These findings based on Figure 9~~ can help to understand the shift in the

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1 observed wintertime maximum of CO₂ between FTIR (January) and NDIR (March-April).
2 The land surfaces of northern hemispheric midlatitudes act as a net CO₂ source during the
3 winter half year, since photosynthesis is largely reduced and respiration and anthropogenic
4 emissions of CO₂ dominate the budget. Hence, the ~~observation of~~ maximum of CO₂ is
5 observed at the end of the winter half year and close to the surface. For the free troposphere
6 above JFJ as observed by the FTIR the direct link to these wintertime releases of CO₂ is
7 weakened due to generally reduced vertical transport. At the same time more frequent
8 transport from and land surface contact in the tropics can be deduced (Figure 9), an area that
9 even during the winter half year may act as a net CO₂ sink due to photosynthetic uptake. An
10 earlier onset of decreasing CO₂ in the free troposphere above JFJ could thereby be explained
11 by different seasonality of transport and vertical mixing. Additionally, the assumption of a
12 fixed a priori CO₂ vertical distribution to retrieve the column integrated CO₂ concentration
13 from the FTIR dataset may contribute partially to the observed shift of ten weeks in the NDIR
14 and FTIR maxima, because it is representing the distribution in winter/spring inadequately.

15 Another hint that the two systems are not measuring the same air parcels can be found in
16 correlation analyses. After omitting outliers, which are mostly caused by synoptic events,
17 thermal uplift of polluted air from surrounding valleys, or other local to regional transport
18 events, the correlation of the two datasets is as large as 0.820, which is quite encouraging
19 considering the different nature of the measurements. By excluding the seasonality from both
20 datasets, the correlation stays almost the same, namely 0.824 but drops to 0.460 if the
21 seasonality is included but the annual CO₂ increase is subtracted. The comparison of the two
22 CO₂ datasets with the annual CO₂ increase and the seasonality subtracted showed a very low
23 correlation of 0.071, which is negligible (Figure 10). Because of possible delays and mixing
24 effects of the CO₂ signal, the time shift as well as the width of the moving average calculated
25 on the hourly values of the NDIR CO₂ values was varied between ± 1440 h and up to ± 600 h,
26 respectively. Shifting the NDIR time relative to the FTIR measurement time creates a ridge of
27 higher correlations around 0 h time shift with a slight tendency towards positive values
28 (Figure 13 A). This ridge-like form is clearly pronounced in the correlation plot between the
29 complete filtered FTIR and NDIR datasets and even more in the datasets without slope
30 (Figure 13 C) than in the correlation of the datasets without seasonality (Figure 13 B). There
31 it is very small and the correlation is high across the whole time shift and averaging width.
32 The constantly high correlation for deseasonalized datasets is due to both datasets containing
33 mostly background air whose CO₂ mole fraction changes are mainly driven by the annual CO₂

1 increase and by the seasonality of the CO₂ signal. Since the larger of the two, the seasonality,
2 is subtracted the high correlation is mainly driven by the slope which was calculated to be the
3 same within uncertainties and stays more or less constant over the examined period.
4 Therefore, the time shift has almost no influence. The remaining fluctuations in the CO₂ mole
5 fractions with higher frequencies than the seasonality seem to play a minor role, because
6 they're almost not visible in the comparison of the datasets without seasonality except for the
7 small ridge (Figure 13 B), or there's no correlation at all, as in the comparison of the two
8 datasets without slope and seasonality (Figure 13 D). This is indicating that the two
9 measurement systems are not measuring the same air parcels, even not with a certain delay, or
10 that the CO₂ signal of the NDIR system which is measured at the lower end of the FTIR
11 column becomes diluted beyond recognition for FTIR by the air mixing processes. The
12 positive effect of the increasing width of the moving average on the correlation is strongest,
13 but still very low, around the first 100 h. Afterwards its main effect is broadening the ridge of
14 the slightly enhanced correlations. The reason for the broadening effect of the increasing
15 width is its smoothing effect on the NDIR values. With increasing width, the influence of a
16 specific NDIR point on the correlation becomes smaller and the NDIR dataset evolves into a
17 smooth sine like curve with decreasing amplitudes, similar to the FTIR dataset, where this
18 form is caused by the higher sampling volume and the dampening due to mixing processes in
19 the atmosphere. However, the small influence of the moving average's width on the
20 correlation means that the correlation of the in-situ and the column measurement is mainly
21 influenced by the slope and the seasonality. Short term fluctuations play a minor role mainly
22 because either their CO₂ signal is dampened too much to be seen in the column measurement
23 or it is not measured at all as e.g. diurnal cycles because of the applied measurement methods.
24

25 **5 Conclusions**

26 Two datasets of CO₂ measurements at the High Altitude Research Station Jungfraujoch in the
27 period 2005 to 2013 were compared. The FTIR system is measuring the attenuation of solar
28 light at different wavelengths caused by molecules of light absorbing gas species in the
29 column between the Sphinx Observatory and the sun. From the obtained spectra, with the
30 knowledge of CO₂ specific extinction bands and the pressure distribution along the path of the
31 light, it is possible to calculate the CO₂ mole fraction in the column. The NDIR system is
32 measuring the CO₂ mole fraction of ambient air at the Sphinx Observatory which corresponds

1 to the lower boundary of the FTIR measurements. The two datasets were filtered with a
2 statistical approach to exclude CO₂ measurements which were influenced by recent transport
3 from the planetary boundary layer. The filtering caused a loss of about 5 % in both, the NDIR
4 and the FTIR dataset.

5 The annual CO₂ increase of the two datasets was calculated with a Monte Carlo approach.
6 Despite an average offset of 13 ppm between the two datasets, which is within the systematic
7 uncertainty affecting the FTIR measurement, the slopes were in good agreement, namely 2.04
8 ± 0.07 ppm yr⁻¹ in the FTIR measurements and 1.97 ± 0.05 ppm yr⁻¹ in the NDIR dataset. The
9 seasonality of the CO₂ signal of the NDIR and the FTIR system is 10.10 ± 0.73 ppm and 4.46
10 ± 1.11 ppm, respectively. The difference is caused by a dampening of the CO₂ signal with
11 increasing altitude due to mixing processes. While the minima of the two datasets both occur
12 in the simultaneously, the maxima of the FTIR dataset was found ten weeks earlier than the
13 NDIR maxima.

14 The difference in the occurrence of the minima is most probably caused by the different
15 transport history of the air masses measured at JFJ and in the column above JFJ. In January,
16 the in-situ system is measuring air from central Europe and the Mediterranean, whereas the
17 air masses of the column measurements are more affected by the subtropic Northern Atlantic.
18 With the onset of spring in Europe, the photosynthetic activity is increasing and the CO₂ mole
19 fraction of air measured by the in-situ system starts to decrease at the end of March. The two
20 filtered datasets as well as the two deseasonalized datasets show a high correlation, whereas
21 the correlation between the two detrended datasets is only mediocre and inexistent in the
22 between the two detrended and deseasonalized datasets. Neither shifting the time of the NDIR
23 measurements relative to the FTIR measurements nor increasing the width of the moving
24 average did increase the correlation between the two datasets significantly. The enhanced
25 correlation values around a time shift of zero indicates that (i) there isn't a systematic time
26 shift apparent and that (ii) the correlation between the two datasets is mainly driven by the
27 annual CO₂ increase and to a lesser degree by the seasonality. Therefore both measurement
28 systems are suitable to measure the annual CO₂ increase, because this signal is well mixed
29 within the atmosphere. Short term variations as the seasonality or daily variations are less or
30 not comparable, because (a) the transport history of the air parcels measured is different, (b)
31 the signal is mixed beyond recognition or (c) since the FTIR has a low vertical sensitivity it
32 was not exploited in the present retrievals and therefore the measured column signal contains
33 mixed information from the troposphere and the stratosphere.

1

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3

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18

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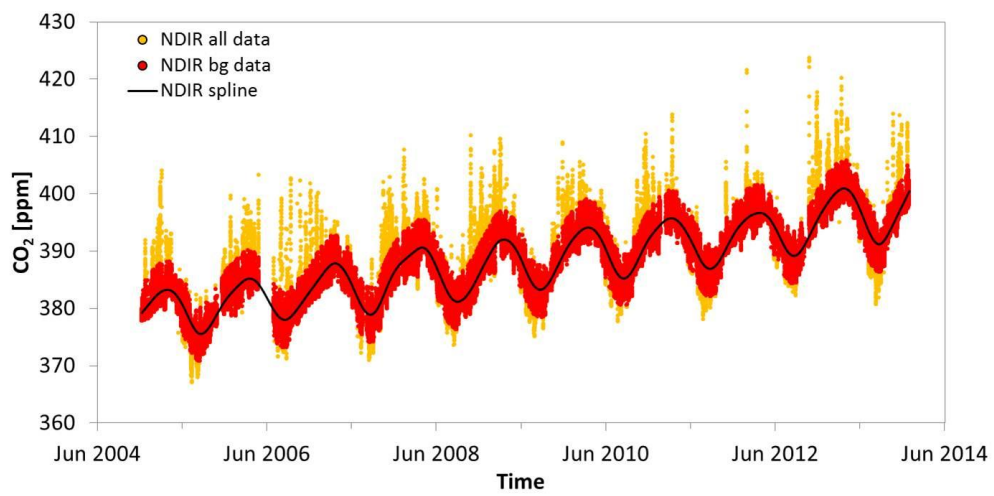
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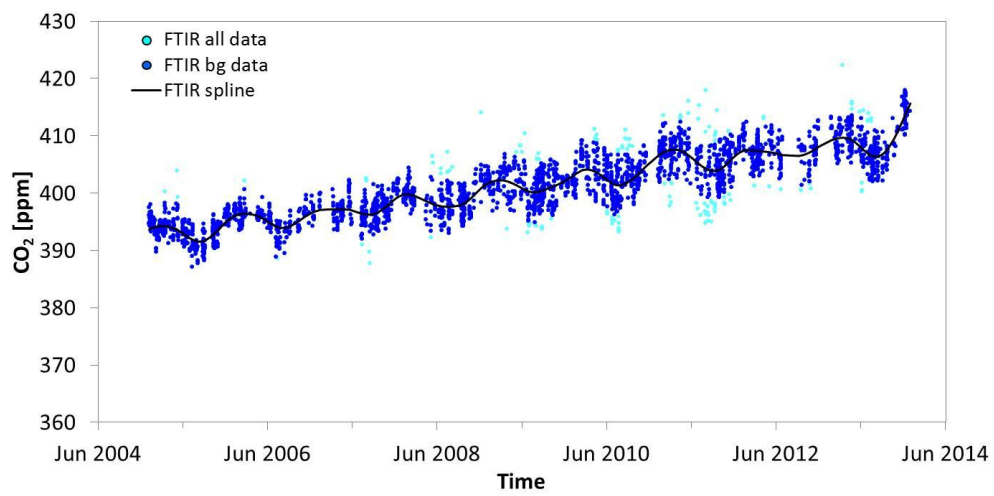
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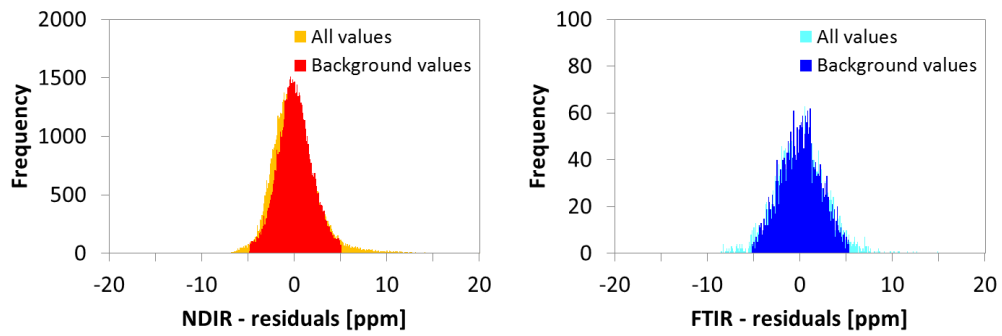
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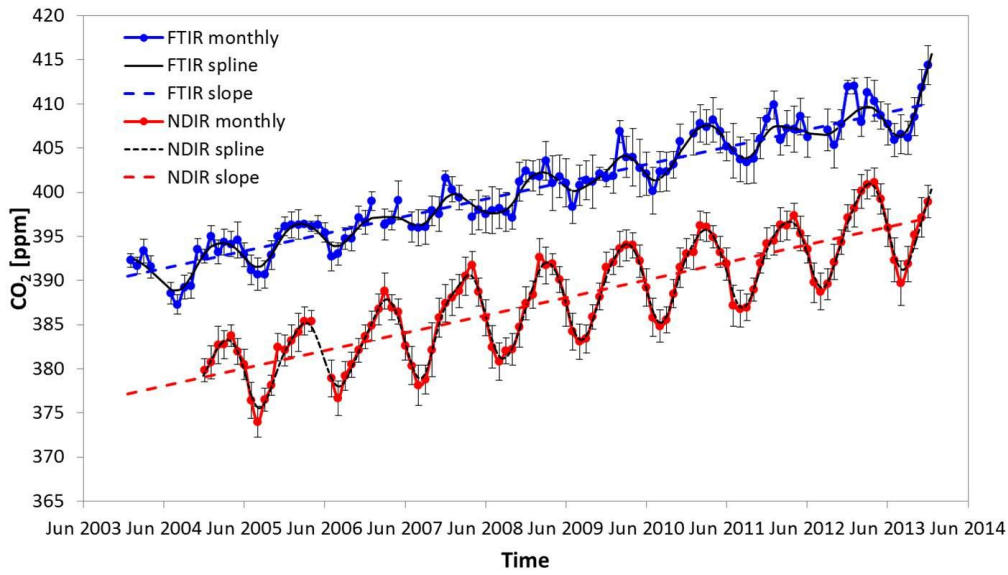
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2 Figure 1. In-situ CO₂ mole fractions of the NDIR measurements as a function of time in ppm
3 at JFJ: All hourly averages before filtering (yellow), hourly averages after filtering (red) and
4 the spline (black line). Note that the yellow points correspond to only about 5 % of the whole
5 dataset.
6



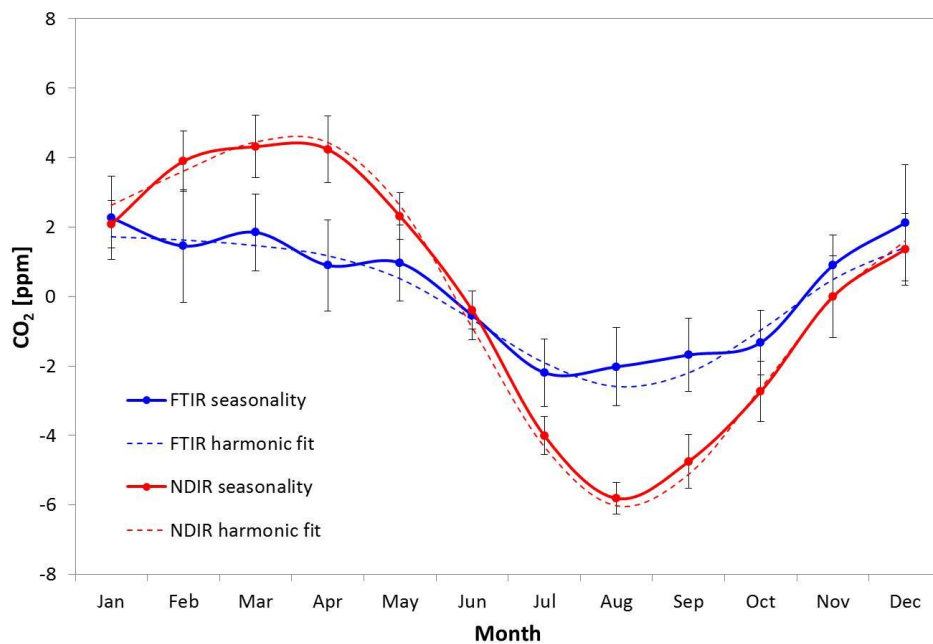
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2 Figure 2. CO₂ mole fractions of the FTIR measurements as a function of time in ppm in the
3 column above JFJ: All hourly averages before filtering (light blue), hourly averages after
4 filtering (dark blue) and the spline (black line). The light blue points correspond to about 5 %
5 of the whole dataset.
6



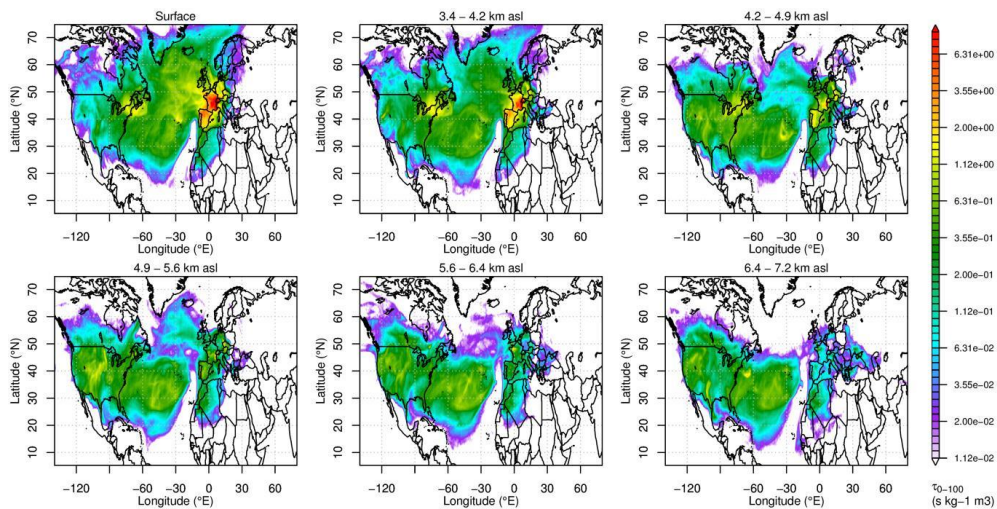
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2 Figure 3. A: Histogram of all NDIR residuals (yellow) and the filtered NDIR residuals
3 representing the background values (red) of the in-situ measurements; B: Histogram of all
4 FTIR residuals (light blue) and the filtered FTIR residuals representing the background values
5 (blue) of the column.
6



1
 2 Figure 4. FTIR and NDIR CO₂ measurements at JFJ as a function of time: Monthly averages
 3 of the filtered FTIR data (blue), spline (black line), the annual CO₂ increase calculated from
 4 the filtered FTIR dataset (blue dashed line), monthly averages of the filtered NDIR data (red),
 5 spline (black dotted line) and the annual CO₂ increase calculated from the filtered NDIR
 6 dataset (red dashed line).
 7

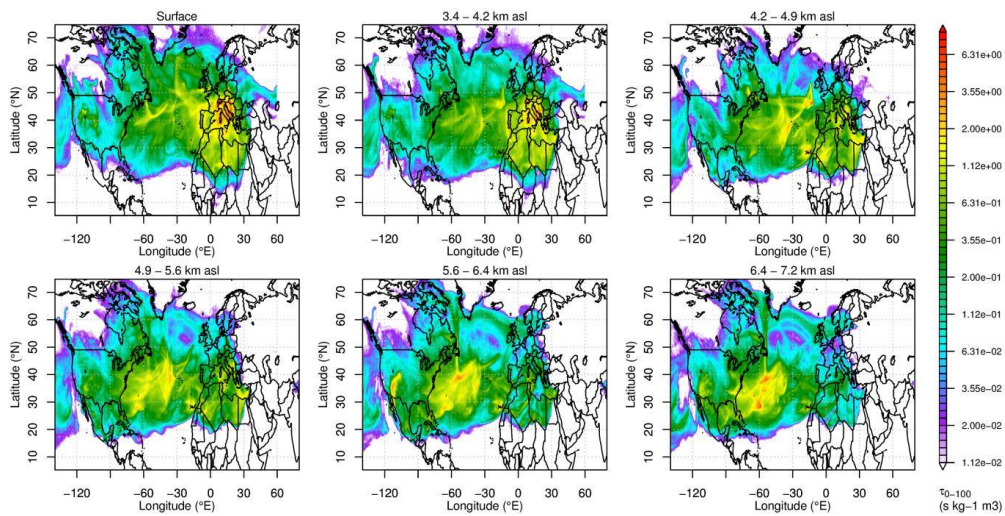


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 2 Figure 5. Monthly averaged seasonality of the filtered FTIR and NDIR CO₂ measurements for
 3 the nine years of the comparison: averaged NDIR seasonality (red), two harmonic fit of the
 4 NDIR seasonality (red dashed line), averaged FTIR seasonality (blue) and two harmonic fit of
 5 the FTIR seasonality (dashed blue line).
 6

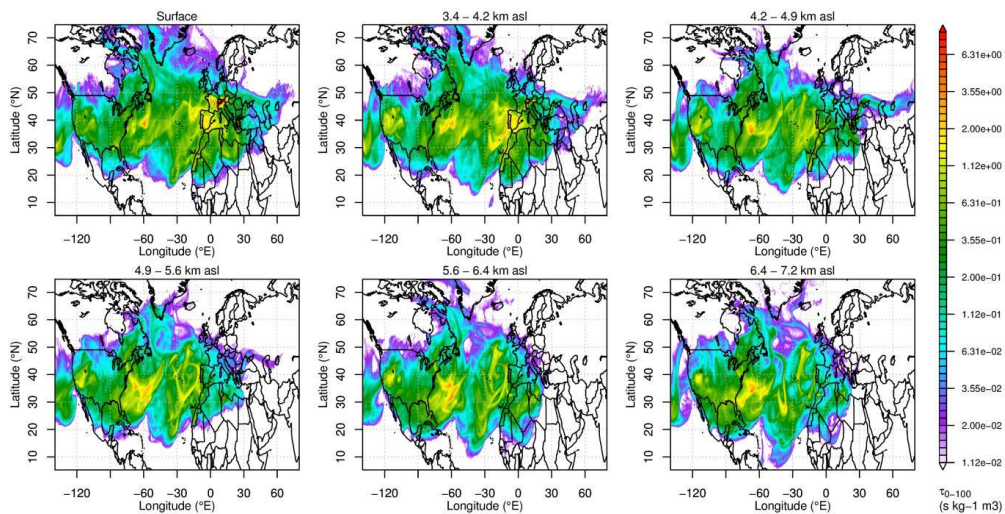


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 2 Figure 6. Surface source sensitivity (footprints) of the air masses at JFJ (surface in-situ) and in
 3 the sub-columns above JFJ in August (CO_2 minimum of FTIR and NDIR time series) in the
 4 period 2009 to 2011 simulated with FLEXPART. The height of the sub-columns is given
 5 above the according subplots, the x-axis is the longitude, the y-axis represents the latitude, the
 6 color code of the sensitivity is given at the right side.

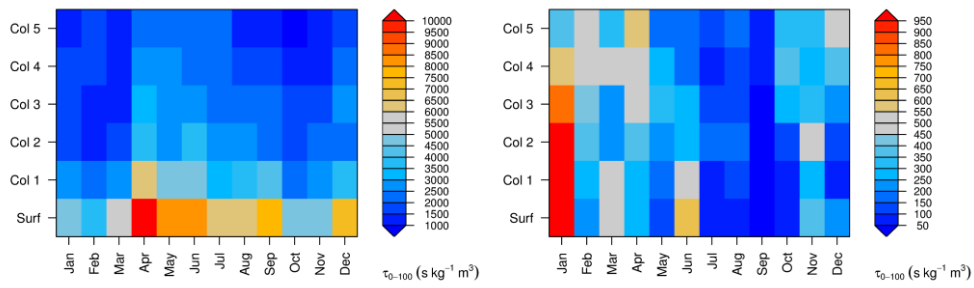
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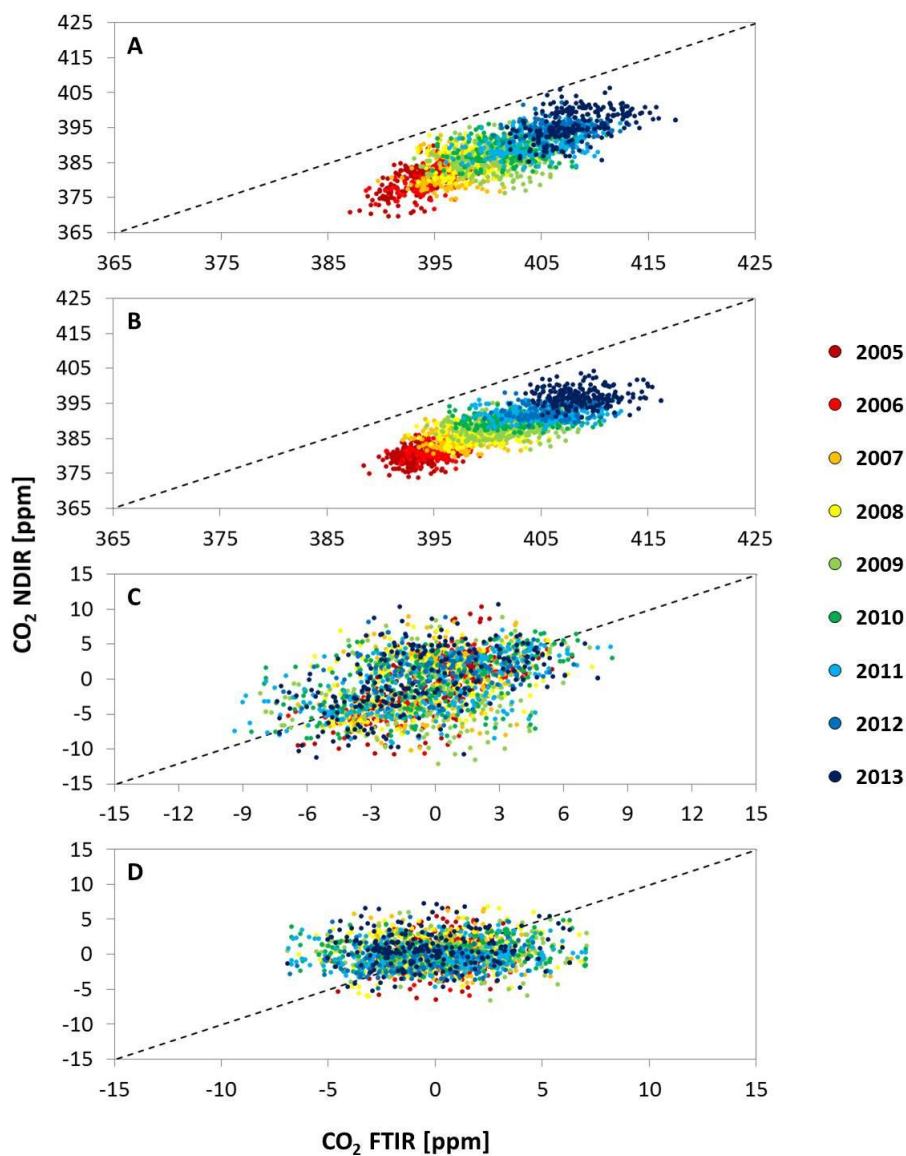
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 2 Figure 7. Surface source sensitivity (footprints) of the air masses at JFJ (surface in-situ) and in
 3 the sub-columns above JFJ in January (CO_2 maximum of the FTIR dataset) in the period 2009
 4 to 2011 simulated with FLEXPART. The height of the sub-columns is given above the
 5 according subplots, the x-axis is the longitude, the y-axis represents the latitude, the color
 6 code of the sensitivity is given at the right side.
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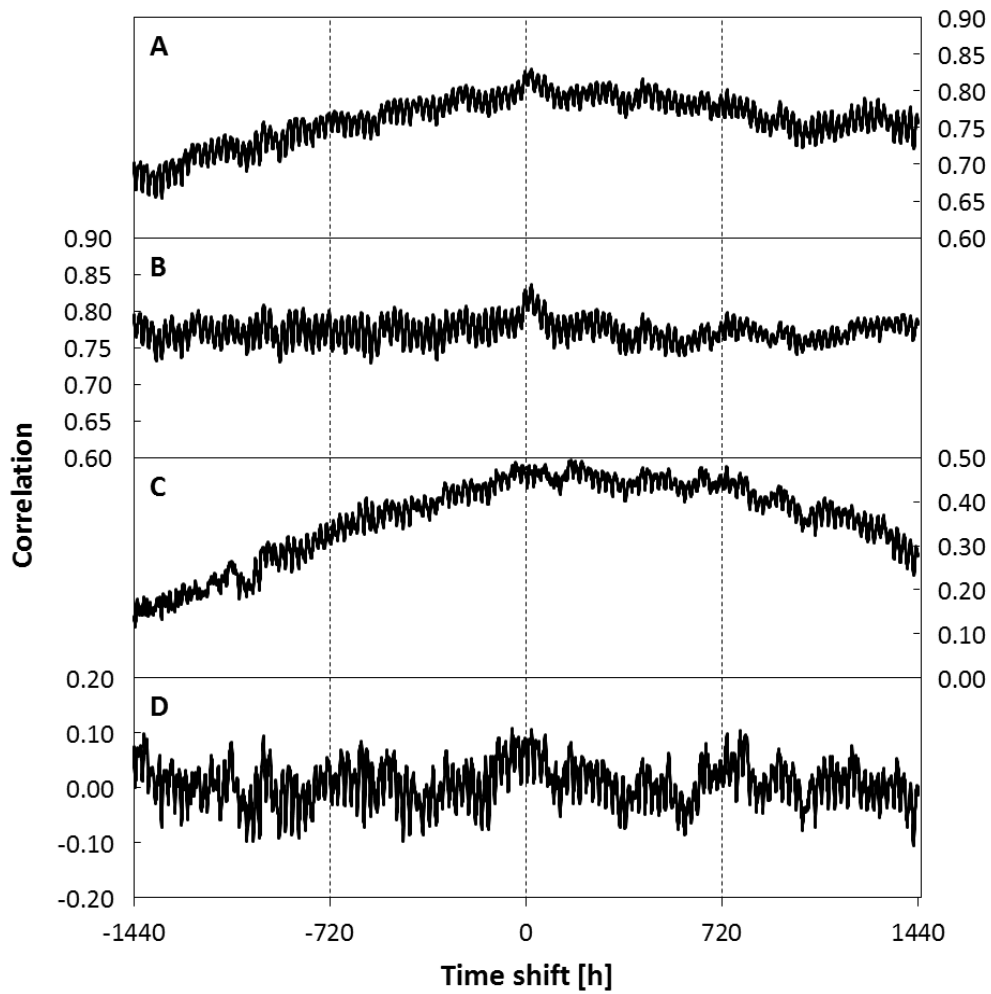
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 2 Figure 8. Surface source sensitivity (footprints) of the air masses at JFJ (surface in-situ) and in
 3 the sub-columns above JFJ in March (CO₂ maximum of the NDIR dataset) in the period 2009
 4 to 2011 simulated with FLEXPART. The height of the sub-columns is given above the
 5 according subplots, the x-axis is the longitude, the y-axis represents the latitude, the color
 6 code of the sensitivity is given at the right side.
 7



1
 2 Figure 9, Annual cycle of FLEXPART derived total surface residence time over land for
 3 different vertical arrival columns above Jungfraujoch: (left) for land surfaces north of 30°N
 4 and (right) for land surfaces south of 30°N.
 5

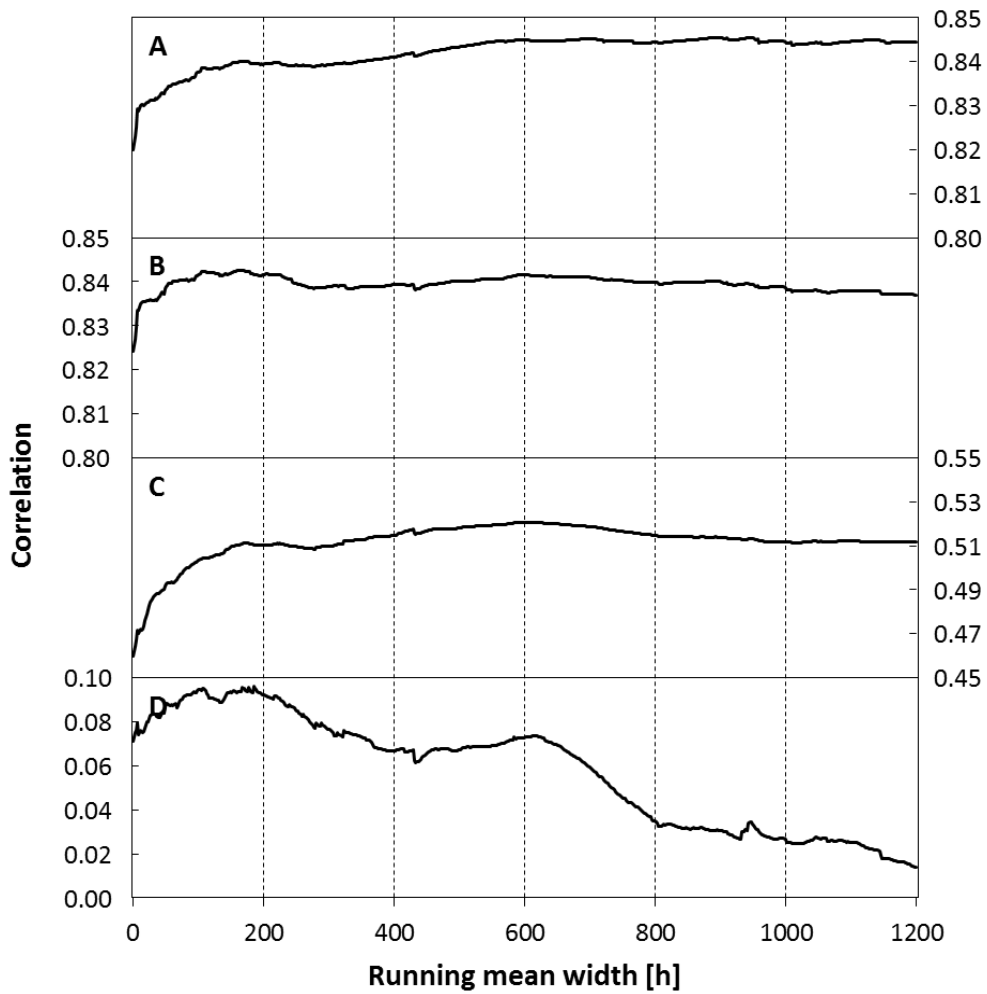


1
 2 Figure 10. Correlation plots of the filtered hourly NDIR CO_2 measurements vs. the filtered
 3 FTIR CO_2 measurements. The different colors refer to the years 2005 to 2013 (see legend). A:
 4 The NDIR CO_2 measurements vs. FTIR CO_2 measurements including both, the annual CO_2
 5 increase and the seasonality; B: As A but without seasonality; C: As A but detrended; D: As
 6 A but with neither annual CO_2 increase nor seasonality. The dashed line is the 1:1 line.
 7



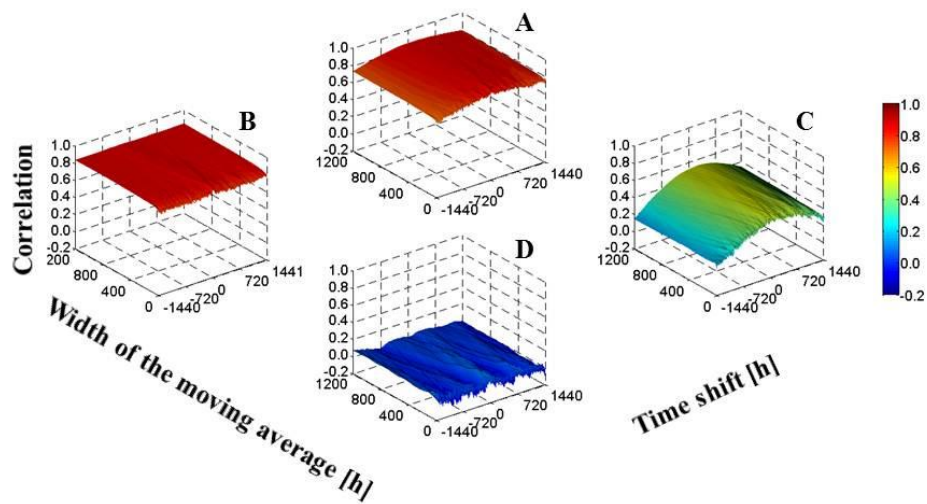
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 2 Figure 11. Evolution of the correlation between the filtered FTIR and NDIR datasets with
 3 changing time shift. A: Correlation between complete datasets; B: Correlation between the
 4 two datasets without seasonality; C: Correlation between the two datasets without trend; D:
 5 Correlation between the two datasets with neither trend nor seasonality.

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 2 Figure 12. Change of the correlation between the filtered FTIR and NDIR datasets with
 3 increasing width of the running mean. A: Correlation between the two datasets with
 4 seasonality and slope; B: Correlation between the two datasets without seasonality; C:
 5 Correlation between the two datasets without slope; D: Correlation between the two datasets
 6 with neither slope nor seasonality.

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 2 Figure 13. Surface plots of the correlation of the NDIR CO₂ measurements vs. the FTIR CO₂
 3 measurements. The x-axis corresponds to the time shift, the y-axis to the width of the moving
 4 average and the z-axis to the correlation between the FTIR and the NDIR dataset, the color
 5 code illustrates the correlation and corresponds to the z-axis values. A: The FTIR CO₂
 6 measurements vs. the corresponding NDIR CO₂ measurements including the annual CO₂
 7 increase as well as the seasonality; B: As A but without seasonality; C: As A but detrended;
 8 D: As A but detrended and deseasonalized.