

Abstract

The University of Bern monitors carbon dioxide (CO₂) and oxygen (O₂) at the High Altitude Research Station Jungfraujoch since the year 2000 by means of flasks sampling and since 2005 using a continuous in situ measurement system. This study investigates the transport of CO₂ and O₂ towards Jungfraujoch using backward trajectories to classify the air masses with respect to their CO₂ and O₂ signatures. By investigating trajectories associated with distinct CO₂ concentrations it is possible to decipher different source and sink areas over Europe. The highest CO₂ concentrations, for example, were observed in winter during pollution episodes when air was transported from Northeastern Europe towards the Alps, or during south Foehn events with rapid uplift of polluted air from Northern Italy, as demonstrated in two case studies.

To study the importance of air-sea exchange for variations in O₂ concentrations at Jungfraujoch the correlation between CO₂ and APO (Atmospheric Potential Oxygen) deviations from a seasonally varying background was analyzed. Anomalous high APO concentrations were clearly associated with air masses originating from the Atlantic Ocean, whereas low APO concentrations were found in air masses advected either from the east from the Eurasian continent in summer, or from the Eastern Mediterranean in winter. Those air masses with low APO in summer were also strongly depleted in CO₂ suggesting a combination of CO₂ uptake by vegetation and O₂ uptake by dry summer soils. Other clusters of points in the APO–CO₂ scatter plot investigated with respect to air mass origin included CO₂ and APO background values and points with regular APO but anomalous CO₂ concentrations. Background values were associated with free tropospheric air masses with little contact with the boundary layer during the last few days, while high or low CO₂ concentrations reflect the various levels of influence of anthropogenic emissions and the biosphere. The pronounced cycles of CO₂ and O₂ exchanges with the biosphere and the ocean cause clusters of points and lead to a seasonal pattern.

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1 Introduction

The present-day concentration of atmospheric carbon dioxide (CO₂) continues to rise. Ongoing emissions of CO₂ and other greenhouse gases will influence the global climate system during the next decades and centuries. It is therefore essential to gain further insight into the processes involving CO₂ exchanges among the atmosphere, the biosphere and the ocean in order to estimate the carbon sources and sinks and to comprehend the global carbon cycle. For that reason greenhouse gases and other atmospheric compounds are constantly monitored at several locations around the world. Long-term records obtained with high precision measurements are used to study the evolution of the changes in time and to budget the sources and sinks of atmospheric CO₂. Atmospheric oxygen observations (often measured as the ratio of oxygen to nitrogen, i.e. O₂/N₂ and expressed as δO₂/N₂ in relation to a standard ratio) can be used as a complementary carbon cycle tracer which provides additional information on the partitioning of anthropogenic CO₂ between the terrestrial biosphere and the ocean (Keeling, 1988; Keeling and Shertz, 1992; Battle et al., 2000; Van Der Laan-Luijkx et al., 2010). Systematic measurements of atmospheric O₂ have begun in 1989 by means of flask sampling at three sites (Keeling and Shertz, 1992). Since then, the flask-sampling network for atmospheric O₂ measurements has been extended over the globe (Bender et al., 2005; Manning and Keeling, 2006; Tohjima et al., 2008) and later on also included continuous on-site measurements (Manning, 2001; Kozlova et al., 2008; Valentino et al., 2008; Popa et al., 2009; Thompson et al., 2009).

The analysis of the correlation between CO₂ and O₂ is especially useful because CO₂ and O₂ are inversely coupled in terrestrial biospheric exchange processes (photosynthesis and respiration) with a rather constant stoichiometric ratio of about -1.1 mol O₂ per mol CO₂ (Severinghaus, 1995). O₂ fluxes between ocean and atmosphere are mainly driven by the biological and the solubility pump, whereas ocean's CO₂ uptake is dominantly driven by the carbonate buffering system. In the air-sea exchange between O₂ and CO₂ due to photosynthesis and respiration of marine biosphere (bi-

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ological pump), the stoichiometric ratio has been determined to be $-1.4 \text{ mol O}_2/\text{mol CO}_2$ (Takahashi et al., 1985) clearly deviating from the terrestrial biospheric ratio. The seasonal variations in the O_2/N_2 ratio of air are partly caused by the warming and cooling of the ocean (solubility pump), also called ocean thermal effect. Typically, O_2 has a low solubility in water and exchanges very fast between the sea surface and the atmosphere with a timescale of a few weeks (Keeling et al., 1993). As oceans temperature rises in spring and summer, gases become less soluble and partly from oceans to the atmosphere. In wintertime, seawater cools and the effect is reversed (Keeling and Shertz 1992; Bender et al., 1996).

On the other hand, during the combustion of fossil fuels O_2 is consumed and CO_2 produced with a ratio of $-1.39 \text{ mol O}_2/\text{mol CO}_2$. This value is the global consummation weighted average of the ratios of the different fuels, like -1.95 for gas fuels, -1.44 for liquid fuels, -1.17 for solid fuels and -1.98 for gas flaring (Marland et al., 2009). Therefore, by knowing the stoichiometric ratios of $\text{O}_2\text{-CO}_2$ exchanges, one can separate the CO_2 uptake into land and ocean components. A sensitive tracer for oceanic air-sea gas exchange is the Atmospheric Potential Oxygen ($\text{APO} = \Delta\text{O}_2/\text{N}_2 + 1.1\Delta\text{CO}_2/0.20946$, with $\Delta\text{CO}_2 = \text{CO}_2 - 365 \text{ ppm}$) as proposed by Stephens et al. (1998). Variations in APO (seasonal and latitudinal differences) are sensitive to CO_2 and O_2 air-sea exchanges plus a residual due to the fossil fuel, but APO is invariant with respect to land biospheric processes (Stephens et al., 1998).

Regarding the fossil fuel influence, our results are compared with the anthropogenic CO_2 emissions distribution in Europe according to the Emission Database for Global Atmospheric Research (EDGAR) emission inventory for the year 2005 (<http://www.pbl.nl/en/themasites/edgar/index.html>). According to the EDGAR emission map (Fig. 1), Germany (especially the Ruhr area), Belgium, the Netherlands, and Poland at the Northern side of the Alps as well as the Po valley (Northern Italy) at the Southern rim of the Alps act as major sources of anthropogenic CO_2 in Europe. It is noteworthy that the Po valley is one of the most well-known source regions for air pollutants observed at Jungfrauoch (Reimann et al., 2004, 2008). These potential source regions are

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the most densely populated and industrialized countries in Europe. An exception in this regard is France due to its high proportion of nuclear energy. The emissions per capita are highest in the Benelux states and also tend to be high in the most northern countries (heating) and Mediterranean countries (cooling systems), while emissions in Russia are only about half of those in Western Europe (Schulze et al., 2009).

One of the most important sites in Europe to monitor atmospheric composition is the High Altitude Research Station Jungfraujoch in the Swiss Alps. Many studies have been carried out at Jungfraujoch on local and long range transport in order to identify source and sink regions of polluted air over Europe reaching this high altitude site (Seibert et al., 1994, 1998; Forrer et al., 2000; Henne et al., 2005a; Cozic et al., 2008; Loov et al., 2008). In this paper, transport of CO₂ and O₂ towards Jungfraujoch is investigated by means of backward trajectories. Trajectories allow tracing the origin of air masses and their potential contact with the surface. Trajectories are mainly used here to classify air masses with distinct O₂ and CO₂ signatures with respect to their origin and hence to characterize the main source and sink regions of these species.

2 Site description, experimental and model methods

2.1 The Jungfraujoch research station

The High Altitude Research Station Jungfraujoch is located on the Southern side of a rocky crest in the Bernese Alps, Switzerland, at an altitude of 3580 m a.s.l. (Sphinx Observatory). It is situated on a saddle between the Jungfrau peak (4158 m a.s.l.) in the west and the Mönch peak (4099 m a.s.l.) in the east. Therefore, the wind is channeled locally in north-south direction. In winter and summer more than 70% of the wind direction is from northwest. North is also the prevailing wind direction in the other seasons.

Due to its location at high elevation and its central position in Europe the site is particularly suited to observe the free tropospheric background above Europe (Schwikowski

et al., 1995; Cozic et al., 2007; Reimann et al., 2008; Lanz et al., 2009). Nevertheless, several studies on aerosol and trace gas composition (Lugauer et al., 1998; Forrer et al., 2000; Coen et al., 2004; Reimann et al., 2004; Li et al., 2005) pointed out that Jungfraujoch is frequently influenced by direct transport from the planetary boundary layer through convection and local thermal wind systems mainly during summer, when vertical transport is more active. In addition to these rather local influences, frontal uplift of air from the European boundary layer followed by quasi-horizontal transport to Jungfraujoch episodically affects the station in all seasons of the year. Therefore greenhouse gas measurements at Jungfraujoch can capture both advected air from the polluted boundary layer as well as air from the lower free troposphere. Moreover, the permanent research station is easily accessible by railway all year round. Local emissions are usually low, since all heating is electrical and waste is transported back to the valley.

The central role of the research station Jungfraujoch is documented by the fact that it was selected as one of the Global Atmosphere Watch stations (GAW) within the framework of WMO activities. In addition, Jungfraujoch is an alpine site within the Network for the Detection of Atmospheric Composition Change (NDACC) and acts as a station within the Swiss national air pollution monitoring network (NABEL) and the European Monitoring and Evaluation Program (EMEP).

2.2 Continuous measurements of CO₂ and O₂ at Jungfraujoch

The Climate and Environmental Physics Division of the University of Bern is monitoring atmospheric CO₂ and O₂ at the High Altitude Research Station Jungfraujoch since 2000 by means of weekly flasks sampling. At the end of 2004 a continuous measurement system for in situ analysis of CO₂ and O₂ was installed in order to get an optimal data coverage to better detect short-term fluctuations in CO₂ and O₂ concentrations. Carbon dioxide is measured by a commercial NDIR infrared analyzer (S710 UNOR, SICK MAIHAK) while oxygen is measured in two ways: paramagnetically, utilizing the paramagnetic properties of oxygen molecules (at the Jungfraujoch Parox 1000

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paramagnetic sensor, from MBE AG, Switzerland is used) and electrochemically, by means of fuel cells. We use Max-250 fuel cells manufactured by Maxtec, USA (Sturm, 2005; Valentino et al., 2007, 2008; Uglietti et al., 2008).

The CO₂ results are reported on the WMO mole fraction scale (XWMO 2007) whereas O₂ values are reported on our internal oxygen scale since no international and common calibration scale exists yet. The difference between the Bern internal scale and SIO scale was set to -556 per meg after measuring several times three standard air cylinders (CA07043, CA07045, CA07047) filled and determined at the Scripps Institution of Oceanography of California, La Jolla.

The continuous measurement system at Jungfraujoch is calibrated using calibration gases contained in 50 L cylinders from CARBAGAS, Switzerland. All standard cylinders are placed horizontally in order to avoid thermal and pressure fractionation of O₂/N₂. It is indeed known that for instance thermal fractionation of O₂ relative to N₂ inside high pressure gas cylinders resulting from small ambient temperature changes can lead to drift in $\delta O_2/N_2$ values for the delivered air on the order of 10–20 per meg over the lifetime of the cylinder (Keeling et al., 1998, 2004, 2005; Langenfelds et al., 2005).

Every calibration gas is passing through the entire system alternatively following an automated sequence written in LabView. We use a high CO₂ calibration gas, which correspond to the low O₂ calibration gas, a low CO₂ calibration gas corresponding to the high O₂ calibration gas and the working gas which should have CO₂ and O₂ concentrations in the range of the air sample values.

The CO₂ measurements are part of the Swiss GCOS (Global Climate Observing System) office program supporting long-term records. Data are archived through the GAW as well as the European project Infrastructure for Measurements of the European Carbon Cycle (IMECC).

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2.3 Trajectory simulations for qualitative allocation of sources and sinks

Trajectory models are a common tool in environmental and atmospheric science to study transport of air masses in the atmosphere. Trajectories describe the pathway of individual air parcels and are typically calculated based on 3-dimensional wind data from a meteorological model (Stohl et al., 1995, 1999; Stohl and Seibert, 1998). Although trajectories describe atmospheric transport only in a simplified way neglecting sub-grid scale turbulent and convective motions, the statistical analysis of a great number of back trajectories from receptor sites has turned out to be a valuable tool to identify sources and sinks of atmospheric trace substances (Scheifinger and Kaiser, 2007).

In this paper a trajectory analysis is performed in order to better explain the measured CO₂ variations and to allocate possible source regions. For this purpose trajectories were calculated using the trajectory model FLEXTRA (Stohl, 2006). The meteorological data for the trajectory calculations were taken from the European Centre for Medium Range Weather Forecasts (ECMWF) at 1° × 1° spatial and 3 h temporal resolution. Trajectories were calculated backwards over five days, once every four hours. Five trajectories were computed for every CO₂ value, a central trajectory ending exactly at the geographical position of Jungfrauoch (7.98° E and 46.55° N), the other four having the endpoint placed in the surrounding of Jungfrauoch, at a distance of 0.25° (about 25 km) in the four cardinal directions (north, south, west and east). By computing more than one trajectory it is possible to obtain an improved statistics of the air mass transport patterns. Hourly means of the measured CO₂ concentrations were first calculated and then averaged over the four hour time intervals each centered on the release time of the five backward trajectories. Trajectories were released from a constant pressure of 700 hPa which roughly corresponds to an altitude of 3000 m. This is lower than the true station altitude but significantly higher than the surface topography in the coarse resolution ECMWF input fields. The choice of 700 hPa was based on tests with different release altitudes where CO emissions of the EMEP emission

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inventory were added to the trajectories passing through the planetary boundary layer and where the corresponding simulated CO concentrations were compared with actual measurements. Information on the altitude of the planetary boundary layer was obtained for all years from ECMWF analyses and forecasts at 3 h resolution.

5 The data analyses cover the period of continuous measurements at Jungfraujoch since the beginning (January 2005) until June 2009.

For the assessment of European source areas the concentration measurements were then combined with “footprints” of air mass contact with the surface calculated from the backward trajectories. These footprints represent the residence time of air masses in the boundary layer over the European continent. Assuming that emissions are uniformly mixed over the depth of the boundary layer, such footprints can be interpreted as potential source sensitivity maps. The total footprint map for Jungfraujoch (Fig. 2) was constructed by using all trajectories ending at Jungfraujoch in the period investigated. Not surprisingly, the influence from the western direction is enhanced in comparison to the east due to the prevailing westerly winds (Reimann et al., 2008). The maximum residence time (scaled to a value of 1) is placed near the end point of all trajectories which is, in our case, Jungfraujoch. The residence times rapidly decrease with increasing distance from Jungfraujoch (approx. with the squared distance) reflecting the rapid decrease in the potential of an emission of unit strength to contribute to the concentrations measured at Jungfraujoch.

10 The footprints also help to understand the general pattern of atmospheric transport to Jungfraujoch. Air masses from the PBL are normally transported to the site by meteorological processes such as fronts, Foehn winds or thermal up-lifting by convection during anticyclonic periods in summer. These events together with associated atmospheric tracer concentrations can be used to assess emission areas from the European continent.

15 20 25 In addition it is possible to investigate particular extreme values in CO₂ concentrations and associate them with corresponding source areas. Anomalous transport associated with such observations is investigated by means of “relative footprints”. These

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are obtained by dividing the footprint for a given set of observations with concentrations above or below a threshold by the total footprint shown in Fig. 2, after first scaling both footprints by the respective number of trajectories. The relative footprint is directly proportional to the Potential Source Contribution Function (PSCF) proposed in other studies (Zeng and Hopke, 1989; Scheifinger and Kaiser, 2007). The only difference is the scaling by the number of trajectories which makes the relative footprint independent of the number of points selected in a given subset. The relative footprint indicates where a selected subset of air masses has spent relatively more (or less) time in the PBL than the “average air mass” observed at Jungfraujoch. The strong radial pattern of absolute footprints with a peak at Jungfraujoch as seen in Fig. 2 is eliminated by the use of relative footprints or PSCF (Scheifinger and Kaiser, 2007).

3 Results

3.1 CO₂ and O₂ time series

CO₂ and O₂ online records from Jungfraujoch are presented in Fig. 3 and the corresponding mean seasonal cycles in Fig. 4. Atmospheric oxygen and carbon dioxide reveal the typical behaviour observed at mid-latitude sites of the Northern Hemisphere. The CO₂ winter maximum is reached in March, when the photosynthetic sink sets in to take up CO₂ and to release O₂. Then the CO₂ mixing ratios decline rapidly during the May–July growing season and reaching a minimum in August. Afterwards, the CO₂ level rises slowly during September–November because of ecosystem respiration and approaches the maximum to start the seasonal cycle again. O₂ is not exactly six-months out of phase with CO₂. It reaches maximal values in the period end of August until the beginning of September (one month later than the CO₂ minimum) and minimal ones in March. This offset in timing of seasonal extremes between CO₂ and O₂ is due to the ocean that affects O₂ stronger than CO₂. Thermal CO₂ outgassing is opposite in sign to the marine photosynthesis and respiration CO₂ flux thus leading to

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only a small seasonal variation in CO₂ air-sea exchange in addition to the dampening effect of the ocean's carbonate chemistry.

On the contrary, the thermal effect and the biological activity act synchronously for the O₂ air-sea exchange, reinforcing the O₂ seasonal cycle (Keeling and Shertz, 1992; Bender et al., 1996). Moreover the O₂ air-sea exchange takes place on shorter times (few weeks) compared to the CO₂ air-sea exchange (1 year) (Keeling et al., 1993).

The ocean is a net source of O₂ at the sea surface during spring and summer, because more organic matter is produced in the euphotic zone than is consumed by respiration. Net O₂ production begins in spring, when phytoplankton has enough light to bloom. Therefore surface seawater becomes supersaturated in O₂ and consequently a flux of O₂ towards the atmosphere is initiated which is reinforced by the thermal outgassing. Part of the organic matter produced during the phytoplankton photosynthesis accumulates in the ocean mixed layer as particulate or dissolved organic carbon (POC or DOC). In fall and winter when the sea surface temperatures cool down, the mixed layer deepens bringing O₂ under-saturated waters to the surface causing a demand of O₂ from the atmosphere and thus decreasing the atmospheric O₂ concentration. This decline continues until the following spring when a new cycle starts (Sverdrup, 1953; Siegel et al., 2002).

The seasonal cycle for the Jungfraujoch in situ measurements (displayed in Fig. 4) was derived by subtracting first the long-term trend (Morimoto, S., et al., 2003) from the background data. The residuals were linearly interpolated to daily values before calculating monthly means for the entire period. The difference between the CO₂ summer minimum and the winter maximum (seasonal amplitude) is 8.7 ± 0.2 ppm, while the opposite variation in the O₂ signal is 80 ± 13 per meg and in APO 23 ± 15 per meg, respectively. These amplitude values are smaller compared to other sites in Europe (CarboEurope atmospheric measurement sites) because Jungfraujoch is at high altitude and the dilution effect plays an important role, diminishing the amplitude. Moreover Jungfraujoch is less influenced by the terrestrial biosphere (especially areas covered by forests) compared to the other remote sites in Europe (i.e. tall towers, marine

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stations).

3.2 Long term and short term variations

The CO₂ and O₂ time series can be separated in three components: First the long term trend covering variations on timescales of years to decades. Second, the mean seasonal component which represents the variations related to the biospheric cycle of photosynthesis and respiration and to a lesser extent to marine CO₂ and O₂ exchanges. Finally a short term component including hourly to weekly variations mainly associated with meteorological variability and corresponding shifts in transport patterns.

The trends were calculated first by subtracting the mean seasonal cycle from the observations and then computing the linear trend for every year. The online record (Fig. 3) is relatively short in time (January 2005–June 2009) to draw conclusive statements about the long term trend. Nevertheless we calculated a mean annual CO₂ growth rate, computed as average of every year's trend as it would be linear, which corresponds to 1.89 ppm/yr. This value is in agreement with the global growth rate (1.91 ppm/yr) calculated by the IPCC for the decade 1995–2005 (Forster et al., 2007).

3.3 Background corrected data

Since our trajectories only account for the recent history (past five days) of the air measured at Jungfraujoch a background corrected record is calculated. The background was computed using an iterative procedure: 30 days running means and their standard deviations were calculated. All data points above and below 2σ (red band in Fig. 3) were then rejected and the procedure was repeated until convergence. The resulting background was then subtracted from the original data producing the background subtracted records of CO₂, O₂ and APO as shown in Fig. 5. This record represents the third, short-term, component of variability mentioned above, and is used in the following to identify the source and sink regions of CO₂ and O₂.

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4 Model experiments

In order to better understand the daily to weekly CO₂ and O₂ variations with regard to their underlying processes and source areas, it is helpful to cross-plot them as done in Fig. 6 (O₂ versus CO₂ (left) and APO versus CO₂ (right)). In the following we focus on the interpretation of specific subsets of points in the scatter diagram: Extreme events of high CO₂ are first investigated in Sect. 4.1 in the form of case studies. A more statistical analysis is then presented in Sect. 4.2 for large clustered subsets of APO versus CO₂. In each case the interpretation of the data is supported by an analysis of air mass origin by means of relative footprint maps.

4.1 Extreme events of high CO₂

A number of particularly high CO₂ concentrations (>10 ppm above average) were recorded at Jungfraujoch as documented in Fig. 5. In the following we focus on two high CO₂ concentration events showing very different relative footprints.

4.1.1 Case 1

The first was observed in the period from 24 to 27 February 2005. According to the in situ measurements CO₂ concentrations began to increase in the morning of 24 February reaching a maximum of 395.75 ppm at around 21:00 UTC and decreased from the morning of 25 February. Then the concentration increased a second time during 27 and 28 February, reaching a maximum concentration of 402.01 ppm. On 25 February the flasks sampling procedure took place in the morning between 06:45 and 07:00 and the mean CO₂ concentration measured afterwards at the University of Bern by mass spectrometry was 393.41 ppm.

The analysis of Black Carbon (BC) at Jungfraujoch, performed by the Laboratory of Atmospheric Chemistry of the Paul Scherrer Institute (PSI), shows a peak of 550.25 ng/m³ on 24 February and another peak of 542.91 ng/m³ on 27 February (data

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available at EBAS Database, <http://ebas.nilu.no>).

Generally, carbonaceous aerosols (BC) are emitted by incomplete combustion processes such as fossil fuel combustion, wood burning, charcoal combustion or natural fires (Stoffyn et al., 1997; Castro et al., 1999). As reported in previous studies (Baltensperger et al., 1991; Petzold et al., 2007) occasionally large aerosol and BC peaks attributed to long-range transport events (e.g., forest fires) occur at Jungfraujoch. In general BC concentrations at Jungfraujoch exhibit a seasonal cycle with lower values in winter and a maximum in summer, due to the injection of air from the more polluted planetary boundary layer during summertime (Weingartner et al., 1999; Henne et al., 2005b).

In the framework of the GAW aerosol monitoring program several measurement campaigns have been conducted at Jungfraujoch in the period 2004–2005 to characterize the physical and chemical properties of tropospheric aerosols. On 25 February the concentrations of the chemical species analyzed (sulfate, nitrate, ammonium, organic matter, BC) were exceptionally high for the winter season and more reminiscent to typical summer values (Cozic et al., 2008).

The relative footprint map (Fig. 7) for the considered period shows high residence time of air masses above the Baltic countries, Eastern Europe and Germany. Actually MeteoSwiss reported for that period an unusual phase of cold weather conditions with repeated surges of arctic air hitting the Alps associated with light snowfall recorded at several weather stations on the Northern side of the Alps. The Southern side of the Alps was protected from these cold fronts and experienced almost no precipitation (Bader and Schlegel, 2005). This persistent weather situation appears to have induced large accumulation of trace gases in these air masses as they moved from the north passing over heavily industrialized countries to reach Jungfraujoch.

4.1.2 Case 2

The second high concentration event discussed here occurred between 21 and 24 November 2007. Also in this case the high values were recorded with both techniques,

i.e. the continuous measurement system and the flask sampling. The maximum concentration, recorded in the morning of 23 November around 08:00 UTC, was 397.4 ppm. The concentration started to increase during the afternoon of 21 November and returned to the mean concentration of 384 ppm in the morning of 24 November.

5 The corresponding relative footprint (Fig. 8, left) shows air masses originating from the Atlantic Ocean and North Africa and passing with high residence times over the Mediterranean Sea. Nevertheless vertical transport to Jungfraujoch during south Foehn events, which is typically associated with precipitation and convection along the southern flank of the Alps, is not well resolved by the ECMWF model at the coarse resolution of only $1^\circ \times 1^\circ$ as used here. We therefore also analyzed trajectories based on
10 much higher resolution ($7 \text{ km} \times 7 \text{ km}$) wind fields of the COSMO model of MeteoSwiss for this day. They reveal rapid vertical ascent from the Po Valley and the Italian peninsula within only one day before reaching Jungfraujoch (not shown).

The high CO_2 concentrations recorded for these days was also associated with relatively high CO values (as observed by the NABEL network). These high trace gas concentrations could be explained by advection from the Po valley due to a typical south Foehn event. However, air reaching measurement sites at the Northern edge of the Alps often originates from altitudes of about 2000 m a.s.l. in the south and not necessarily from low altitude in the boundary layer (Seibert, 1990; Campana et al., 2005).
20 Consequently, there are south Foehn events which transport highly polluted air from the Po Basin to the Jungfraujoch and others with no sign of pollution when the origin was probably above the boundary layer (Campana et al., 2005).

In this study case it is very likely that the high CO_2 concentrations are caused by contact of these air masses with polluted air above the Po valley before arriving at the Jungfraujoch (Forrer et al., 2000) despite rather short residence times over the Po valley.
25

Indeed Meteo Swiss (Fig. 8, right) reported for those days (21–23 November 2007) the formation of a depression over the Bay of Biscay which has sparked an inflow from the southwest and south with the addition of moist air into the Southern rim of the

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Alps. This has established a classical Föhn situation with substantial precipitation amounts on the Southern side of the Alps (Bader and Croci-Maspoli, 2007). Moreover the analysis of wind speed and directions recorded at Jungfraujoch by Meteo Swiss confirms the south flow showing that the wind direction changed abruptly on the 18 from north to south until 25 of November, when the wind direction changed back to north.

In contrast to case 1 the BC measurements at Jungfraujoch did not show any anomalous values during this event. It is likely that, in contrast to CO_2 and CO , the aerosols have been washed out by precipitation. This increase of CO_2 without a simultaneous increase of BC may thus be interpreted as a typical indicator of south Föhn events because during such events aerosols tend to be washed out by precipitation.

4.2 Origin of air masses for different CO_2 and APO concentrations subsets

This section investigates the influence of different air mass origins on CO_2 and O_2 concentrations at Jungfraujoch in a more statistical way, by analyzing large numbers of trajectories associated with specific clusters of points in the APO– CO_2 scatter plot. Each cluster is represented by a certain range of CO_2 and APO values. Note that the advantage of using APO instead of O_2 is that no rotation of the corresponding selection boxes is required. In a scatter plot of O_2 versus CO_2 a rotation would be necessary to align the boxes with the general negative slope of the O_2 versus CO_2 correlation (see Fig. 6, left panel).

The selection of clusters is illustrated in Fig. 9, left panel. We only select subsets of points with outstanding characteristics. The selection is somewhat subjective, and it is a compromise between representing clear (outstanding) signals in trace gas concentrations and including a sufficiently large number of points to obtain robust patterns in the corresponding trajectory footprints.

The box in the middle (Fig. 9, left panel, subset 1) corresponds to background values. All points are within the 2σ bands of CO_2 (± 2.71 ppm) as shown in Fig. 3 (red band) and APO (± 37 per meg). The other five clusters are defined as follows: points with

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enhanced CO₂ and reduced APO (Fig. 9, left panel, subset 2); points with negative CO₂ and negative APO (subset 3); points with negative CO₂ but enhanced APO (subset 4); points with high APO but normal CO₂ (subset 5); points with regular CO₂ and low APO (subset 6). In the following all selected clusters defined above are discussed.

The occurrence of specific clusters follows a seasonal cycle as indicated in Fig. 9, right panel. Air masses enhanced in CO₂ are preferentially observed in winter, those depleted in CO₂ in summer. Due to the seasonal cycle in air-sea exchange there is also a seasonal cycle in APO with a tendency of negative anomalies to occur in spring and positive anomalies in autumn.

4.2.1 The background cluster (subset 1)

By applying the back trajectories analysis to background values (all data within $\pm 2\sigma$), the relative footprint map shown in Fig. 10 is obtained. Values below 1 dominate (blue colours), indicating that these air masses had less than average contact with the European boundary layer. This demonstrates that the trajectory classification is very successful for this class. There seem to be somewhat more air masses classified as background in both spring and autumn (mainly October), possibly because these are transition seasons for the biosphere with neither large emissions nor uptake. There is also a north-south gradient as documented by the colours. Background air masses from the north tend to have been even less in contact with the PBL than those from the south. This is understandable from a meteorological point of view since advection of background air from the north is typically associated with anticyclonic flow and therefore subsidence. Such conditions appear to be ideal to observe undisturbed free tropospheric air.

4.2.2 Enhanced CO₂ and reduced APO and O₂ (subset 2)

The high CO₂ and low O₂ concentrations recorded at Jungfraujoch occur generally in wintertime because of increases due to the lower biospheric activity (more O₂ is used

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for respiration) and higher anthropogenic CO₂ emissions in Europe due to the burning of different fossil fuels (solid, liquid or gas) (Rotty, 1987), thus enhancing the decrease of atmospheric oxygen. In wintertime the boundary layer is mostly located below the altitude of Jungfrauoch and the high CO₂ concentrations cannot be explained by thermal uplift of polluted air masses from the valleys surrounding Jungfrauoch. These high concentrations are thus likely associated with air masses which had resided in the polluted boundary layer for extended periods of time and were then transported to the free troposphere probably in connection with frontal activity. The left panel of Fig. 11 represents all CO₂ pollution events which mostly but not exclusively occurred between October and April. Since the average PBL residence time of air masses reaching Jungfrauoch is generally about 20% lower in winter than in summer a winter only footprint map was calculated (Fig. 11, right). This figure emphasizes the enhanced residence times of these air masses over the eastern parts of Europe (Poland and Germany). Thus the high CO₂ concentrations may be explained by the passage of air masses above polluted areas in those countries. Therefore, from the EDGAR emission map (Fig. 1) these areas can be considered as potential source regions for polluted air masses reaching Jungfrauoch. The elongated, curved patterns of enhanced residence times (yellow to red areas in Fig. 11, right) suggest that transport associated with such events typically followed a cyclonic path supporting the hypothesis of the importance of frontal lifting. In both case studies presented earlier, the upward transport to Jungfrauoch was indeed connected with fronts.

4.2.3 Negative CO₂ and negative APO (subset 3)

Air masses with low CO₂ and comparatively low O₂ are almost exclusively observed in summer and describe air masses advected from Eastern Europe (Fig. 12). The low CO₂ concentration is an effect of plant CO₂ uptake, but O₂ in these air masses is not enhanced as would be expected from photosynthesis. Therefore, a strong oxygen sink is needed. To our knowledge four options may be considered for this sink: (i) fossil fuel consumption, (ii) stratosphere-troposphere exchange (STE), (iii) atmosphere-

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ocean exchange and (iv) oxidation of soil organic material. Option (i) is rated as improbable considering that the percentage of fossil fuel emissions (Fig. 1) compared to the natural carbon exchange is low its influence on the oxygen budget is limited despite the higher carbon oxidation ratio ($-1.39 \text{ mol O}_2/\text{mol CO}_2$). To option (ii), it has recently been reported that O_2/N_2 ratios are enriched with increasing altitude (Ishidoya et al., 2008) associated with decreased CO_2 concentrations (Gurk et al., 2008). This behaviour is explained by altitude dependent air age. Hence a reduced STE would lead to an apparent O_2 sink and CO_2 source due to delayed upward transport of ground-based emission signals. If this option would be the major cause then ozone measured at Jungfraujoch should express lower values as reaction to this STE reduction, yet ozone shows a summer maximum. Therefore, this option seems also improbable. Option (iii): Atmosphere-ocean exchange is important for atmospheric O_2 concentrations. In summer, however, we would expect increased outgassing from the ocean due the increased temperature. This is exactly opposite (neglecting the marine biosphere activity) to what is needed. Furthermore, air masses for subset 3 are dominantly originating over the continent with an actively growing biosphere (low CO_2 , high O_2). Yet, a slight timing difference of the temperature induced ocean source for O_2 in summer compared to the terrestrial biosphere O_2 source could lead to an ocean-continent gradient. The observed low CO_2 , O_2 and APO values for subset 3 might be the result of such a gradient with advected air masses from the continent that have not been in contact with the ocean surface for an extended period of time (weeks) and therefore have O_2 (APO) values which are lagging behind the normal seasonal cycle observed at Jungfraujoch (Fig. 4). Note that there is a significant increase in O_2 as well as APO from June to August such that this delay would indeed lead to anomalously low APO making this option highly probable. Finally, option (iv) might be acting in parallel since dry summer soils consume more oxygen in order to oxidise soil organic material or released methane from wetlands than it is produced from photosynthesis (Pezeshki and Delaune, 2002; Cuna, et al., 2008). Whether this option can play a major role is difficult to judge due to the sparse information about this process of soil oxidation.

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4.2.4 Negative CO₂ and positive APO (subset 4)

The average CO₂ background concentration decreases during summertime due to photosynthesis whereas the background O₂ is high (ocean outgassing, land and ocean biosphere contribution). This effect is further highlighted by the background corrected data in CO₂ whereas the positive APO values (accounts for the land biosphere influence by its definition) is consistent with increased ocean outgassing. The footprint map generated from this cluster (Fig. 13) consists predominantly of summer trajectories and represent air masses originating from the North Atlantic Ocean and additionally having high residence time over Scandinavia. This is in line with the fact that (i) Scandinavia has low fossil fuel CO₂ emissions (as shown by the Edgar emission map, Fig. 1) but significant plant/forest-covered areas and that (ii) the ocean outgassing is increased during summer in the North Atlantic.

4.2.5 High APO (corresponding to high O₂) and normal CO₂ (subset 5)

The subset with regular CO₂ and high APO values (corresponding also to high O₂ values, subset 5) is associated with air masses advected from the Western North Atlantic Ocean, in particular during winter and spring (from November to April). The corresponding relative footprint map is shown in Fig. 14.

To support our view of marine air masses being responsible for high APO (and high O₂) concentrations at Jungfraujoch, we looked at the monthly maps of chlorophyll-*a* for the investigated period (January 2005–June 2009) available from the GlobColour project (<http://www.globcolour.info/>). The regions around Europe with highest marine photosynthesis rates are the Baltic Sea and the Atlantic Ocean in the vicinity of the coasts of Mauritania and Morocco. The maximum concentration of chlorophyll-*a* along the Western African coast is around 5–10 mg/m³ and occurs in general between February and April. Therefore, it can be argued that the high O₂ concentrations measured at Jungfraujoch could be influenced by the marine biospheric activity especially in the Atlantic Ocean and particularly in spring as shown in the footprint map.

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4.2.6 Regular CO₂ and low APO (subset 6)

Most of the events associated with regular CO₂ and low O₂ (low APO) concentrations were observed in wintertime. The relative footprint map displayed in Fig. 15 shows a relative high residence time of air masses over the Mediterranean Sea and over Northern Africa. A possible mechanism for our observation could be an uptake of atmospheric oxygen by up-welling waters in the (Eastern) Mediterranean Sea under-saturated in oxygen (Kress and Herut, 2001). This is in line with a deepening of the thermocline during winter time bringing deeper waters to the surface. Indications that such O₂ uptake might be occurring at least periodically is given in a recent publication (Van Der Laan-Luijkx et al., 2010).

5 Conclusions

A trajectory analysis was used to identify and classify carbon dioxide and oxygen observations at Jungfraujoch. In this study, 5-days backwards FLEXTRA trajectories were computed using meteorological data from ECMWF. Trajectory studies for different CO₂ concentration situations allow us to decipher different source and sink areas over Europe. In general, trajectories associated with particularly high CO₂ concentrations show transport of air masses from North Eastern Europe (Poland and Germany) or passing over areas like the densely populated Ruhr area in Germany as well as the Po Valley in Northern Italy both often heavily loaded with fossil fuel CO₂. Transport from the Po Valley is mainly connected to south Foehn events.

For a detailed footprint analysis, data were subdivided into six clusters using scatterplots of background corrected APO versus CO₂ concentrations. These selections led to convincing relative footprints for background levels (subset 1) and those that are either influenced by pollution (subset 2), terrestrial biosphere (subset 3) or ocean outgassing (subset 4) and ocean circulation processes (subset 5 and 6).

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All data within $\pm 2\sigma$ are included in the background subset. The relative footprint map generated from this cluster shows that transport of air masses classified as background occurs mainly during spring and autumn, when transition between the biospheric large uptake (of CO_2 starting in spring and of O_2 in autumn) and release (beginning in spring-time for O_2 and in autumn for CO_2) occurs. Moreover background air masses from the north, typically associated with anticyclonic flow and subsidence, tend to have been less in contact with the PBL than those from the south. Therefore these conditions are ideal to observe undisturbed free tropospheric air.

The second cluster of points in the APO versus CO_2 scatter plot corresponds to enhanced CO_2 and reduced APO and O_2 . The high CO_2 concentrations are likely associated with winter air masses having enhanced residence time over the eastern parts of Europe (Poland and Germany) and transported to the free troposphere probably in connection with frontal lifting.

Subset 3 describes predominantly summer air masses advected from Eastern Europe. Vegetation uptake explains the low CO_2 concentrations of this cluster as expected from photosynthesis, but not the low O_2 . Despite the higher carbon oxidation ratio for fossil fuel combustion, the influence of the fossil fuel emissions on the atmospheric oxygen budget is limited compared to the biospheric carbon exchange. Therefore, an additional O_2 sink has to be taken into account. The air masses associated with this cluster are mainly originating over the continent. Therefore, it might be that dry summer soils consume more oxygen in order to oxidise soil organic material than it is produced from photosynthesis.

The fourth cluster of points shows negative CO_2 and positive APO. The corresponding footprint map represents predominantly summer air masses originating from the North Atlantic Ocean and additionally having high residence time over Scandinavia which has significant plant/forest-covered areas and low fossil fuel CO_2 emissions, thus explaining the low CO_2 concentrations. Moreover the ocean outgassing increases during summer in the North Atlantic due to its biological activity.

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Cluster 5 has high APO (corresponding to high O_2) and regular CO_2 and it is associated with air masses advected from the Western North Atlantic Ocean, in particular during winter and spring. The corresponding footprint map exhibits the main provenance of air masses from the ocean, thus supporting the view of marine biospheric activity being responsible for high APO and oxygen concentrations measured at Jungfraujoch.

The last subset (6) represents events observed in wintertime and associated with regular CO_2 and low O_2 (low APO) concentrations. The associated air masses have high residence time over the Mediterranean Sea and the low atmospheric O_2 could be explained by an uptake of oxygen from uplifted under-saturated sea water.

Finally, this study demonstrates the potential of trajectory analyses for classifying air masses and thus explaining particular short-term features of the CO_2 and O_2 records. The addition of more atmospheric trace gases, such N_2O , CO and CH_4 would further constrain the influence of air mass transport on measured concentrations. To better resolve the transport, it would be desirable to use a Lagrangian particle dispersion model rather than a simple trajectory model and to include wind fields at local scale to better resolve the flow pattern over the complex topography of the Alps.

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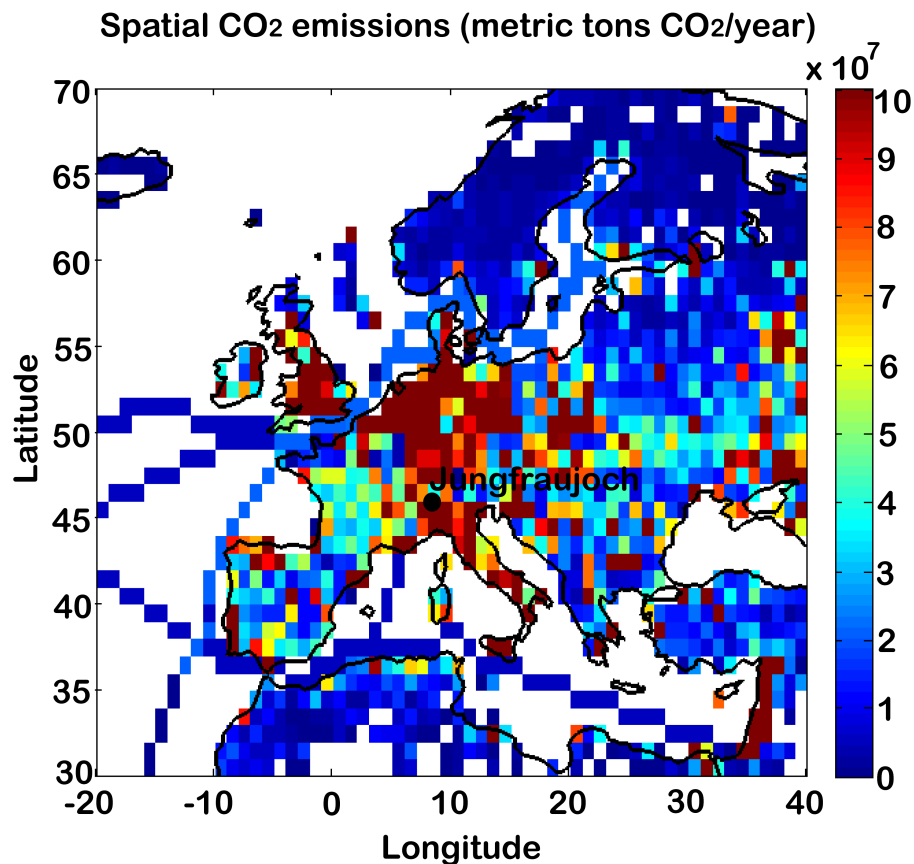


Fig. 1. EDGAR 3.2 Fast Track 2000 dataset (32FT2000) comprises anthropogenic CO₂ emissions for the year 2005 (<http://www.pbl.nl/en/themasites/edgar/index.html>). The colours represent the metric tons of CO₂ emitted by the European countries per grid cell. The strategically good position of Jungfraujoch to observe European CO₂ sources is displayed as a black dot.

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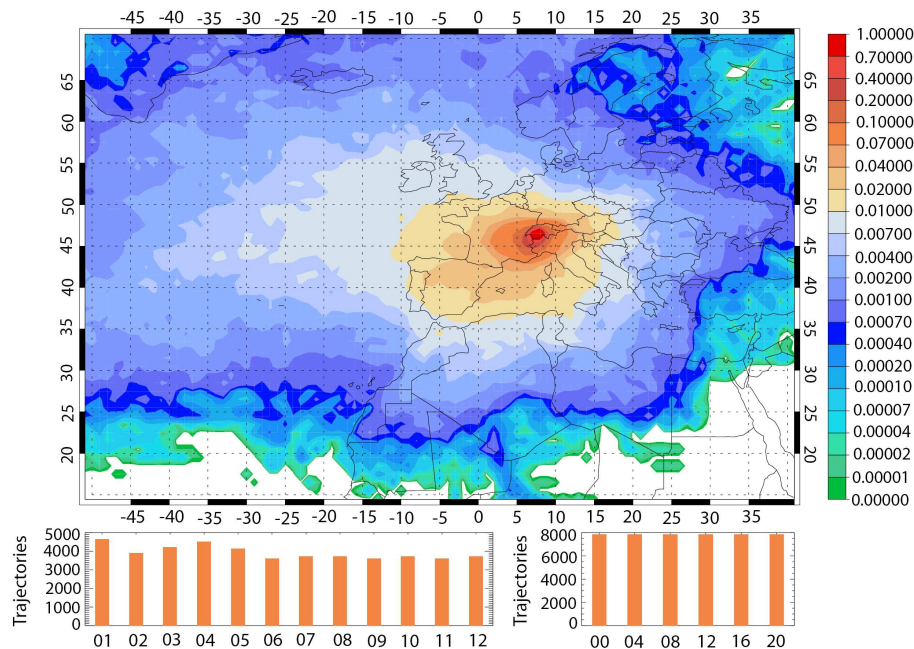


Fig. 2. Total footprint of all air masses sampled at Jungfraujoch during the investigation period (2005–2009): Colour contours represent residence times of air parcels within a specific geographic grid cell relative to the maximum residence time. The maximum residence time corresponds to the end point of all trajectories, i.e., to Jungfraujoch. The lower panels show the number of trajectories per month (on the left) and per hour of the day (on the right) used to create the footprint map. Since trajectories were only calculated for valid observations, these panels demonstrate the rather uniform sampling of CO₂ over the different months of the year and hours of the day.

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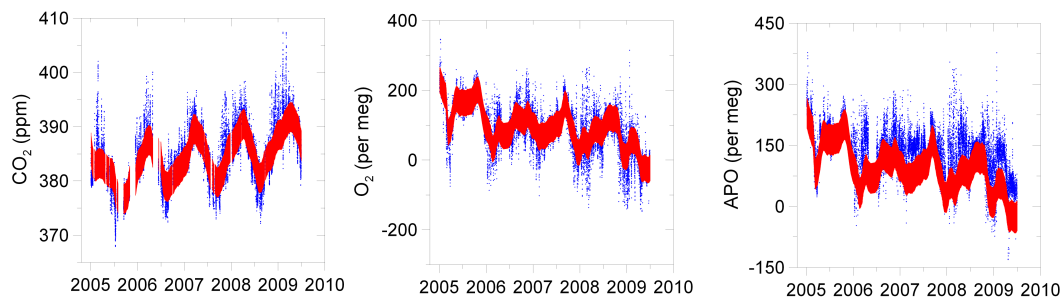


Fig. 3. CO₂ (left), O₂ (centre) and APO (right) hourly means (in blue) with the iteratively calculated background range (in red). After background subtraction, the remaining data is considered and included in the trajectory model.

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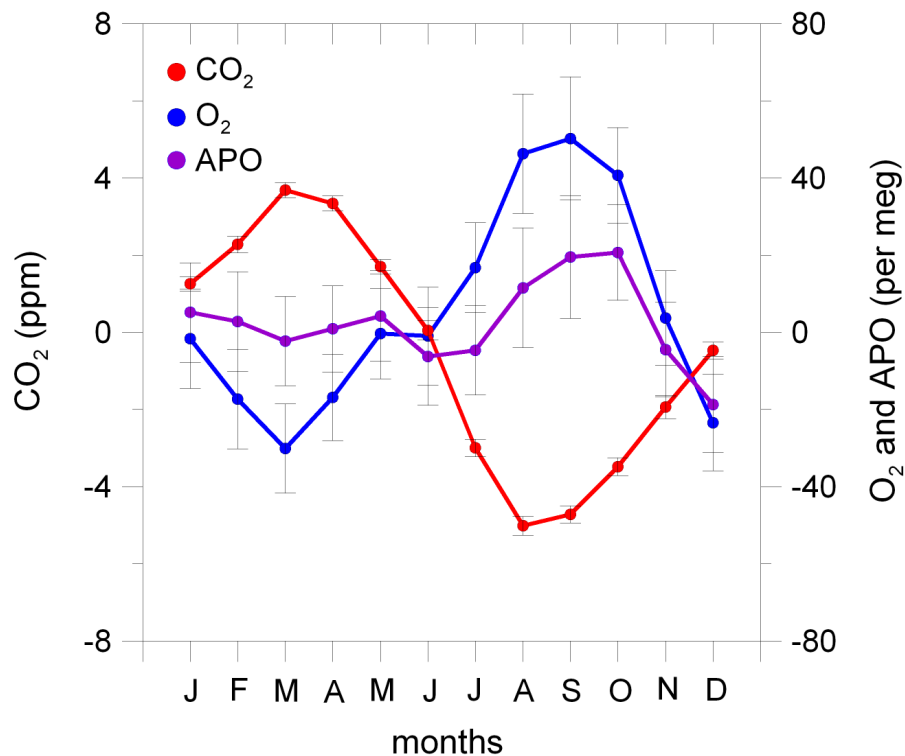


Fig. 4. Seasonal cycle of CO₂ (red curve), O₂ (blue curve) and APO (purple curve) at Jungfraujoch expressed as deviation from the mean concentration. The seasonal cycle of O₂ is approximately six months out of phase with the CO₂ cycle. APO is slightly decreasing from January until June followed by a maximum in autumn. Error bars are the measurement uncertainties.

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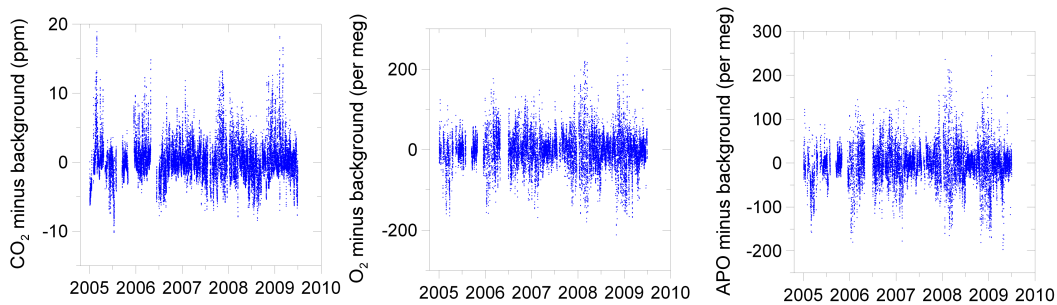


Fig. 5. CO₂ (left), O₂ (center) and APO (right) records after background subtraction.

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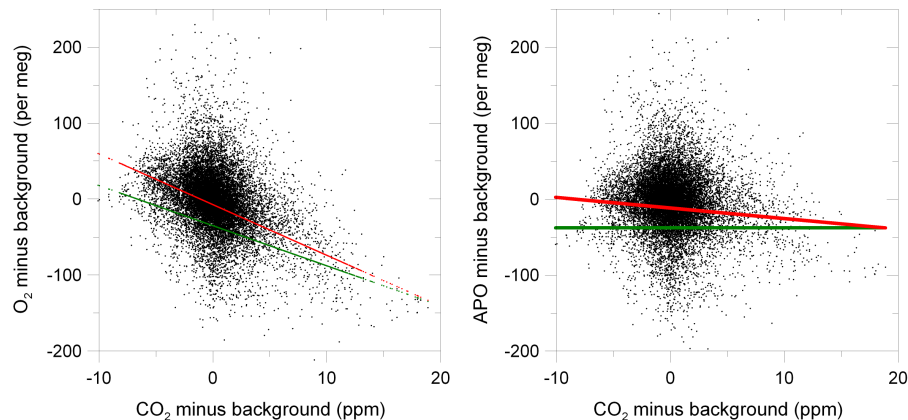


Fig. 6. Scatter plot of O_2 versus CO_2 (left) and of APO versus CO_2 (right). Both records are background corrected. The slopes of fossil fuel burning (-1.4 mol/mol , red line) and of land photosynthesis and respiration (-1.1 mol/mol , green line) are overlaid for reference. APO is invariant with respect to land biospheric processes by its definition (green line, right panel).

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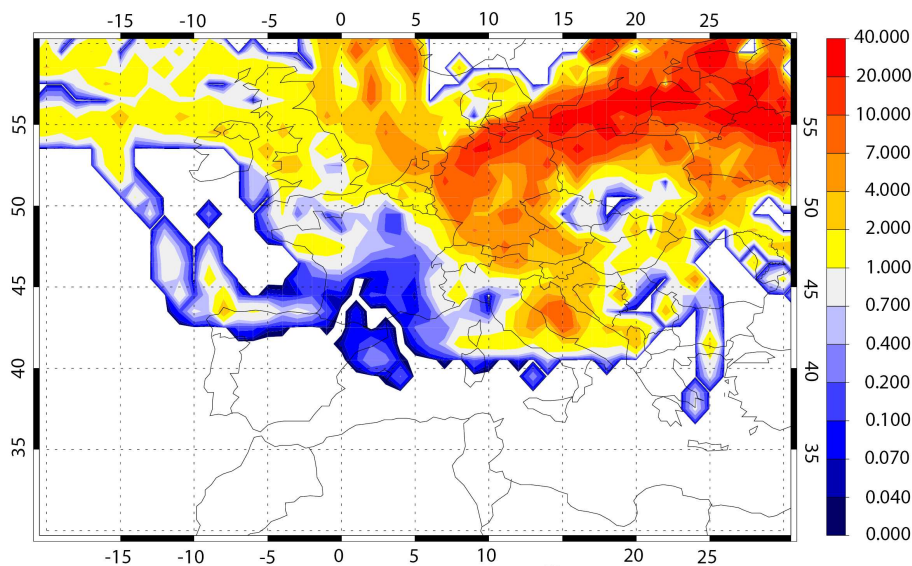


Fig. 7. South Foehn situation: relative footprint map of trajectories associated with exceptionally high CO₂ concentrations recorded with the continuous measurements system at Jungfraujoch between 24 and 27 February 2005.

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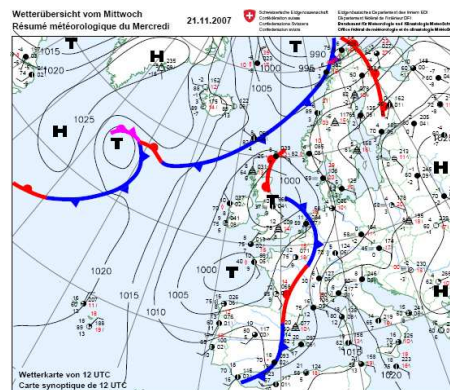
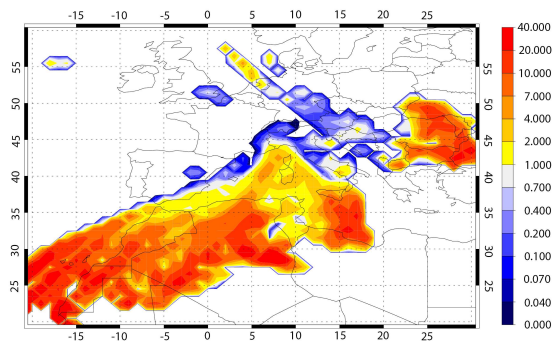


Fig. 8. Relative footprint map of trajectories corresponding to high CO_2 concentration measured on 23 November 2007 (left). South Foehn situation drawn by Meteo Swiss (right, meteorological situation of the 21 November).

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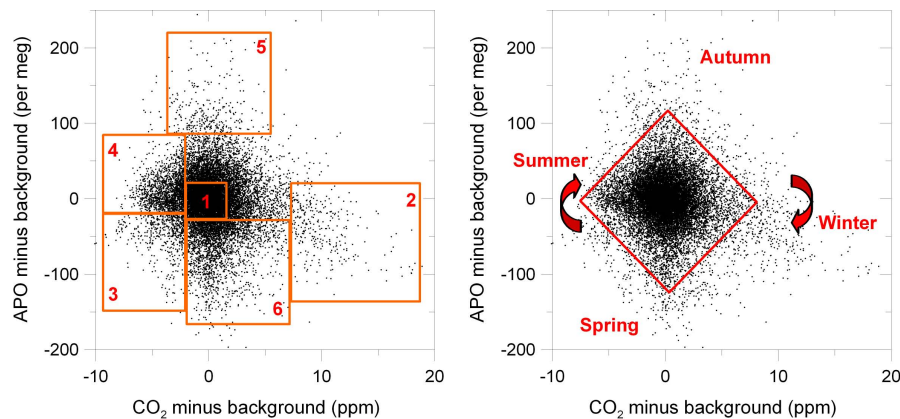


Fig. 9. Different selections (see text for explanations) in the scatter plot of the background corrected APO versus background corrected CO₂ (left panel). Seasonal cycle of defined clusters (right panel).

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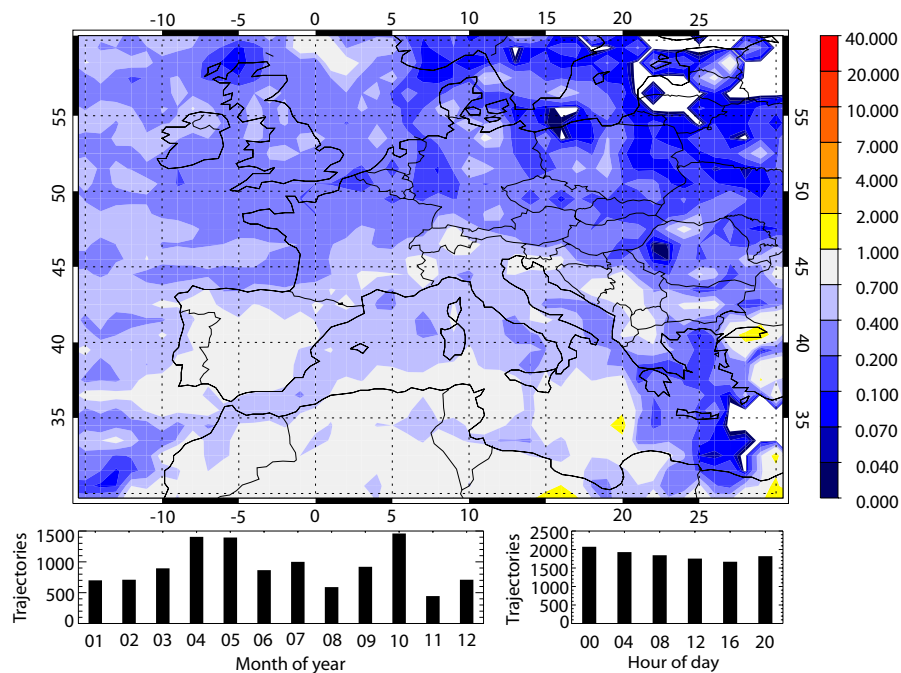


Fig. 10. Relative footprint map of trajectories associated with background values of CO_2 and O_2 recorded at Jungfraujoch (background calculated as explained in Sect. 3.1).

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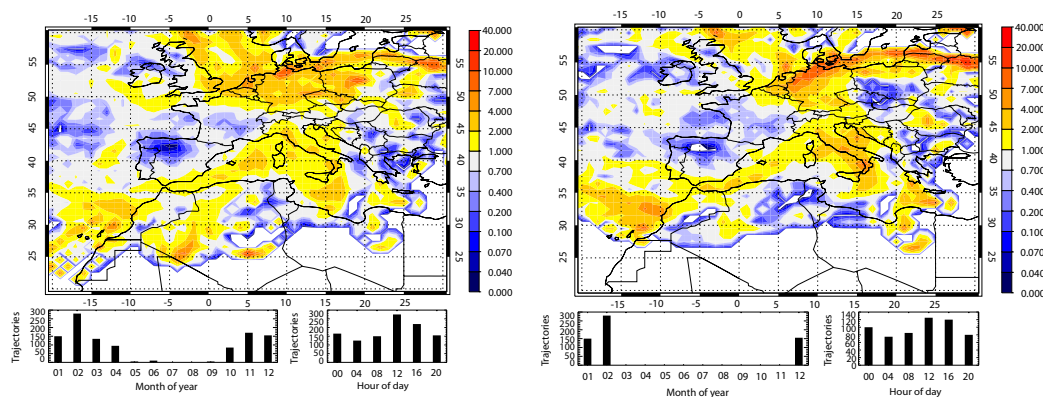


Fig. 11. Relative footprint maps for the situation with high CO_2 and low O_2 . The left panel represents the relative footprint map for year round values whereas the right panel shows the winter only selection. The trajectories are mainly coming from Northeastern Europe with increased residence times over Northern Germany and Eastern European Countries (yellowish and reddish colours).

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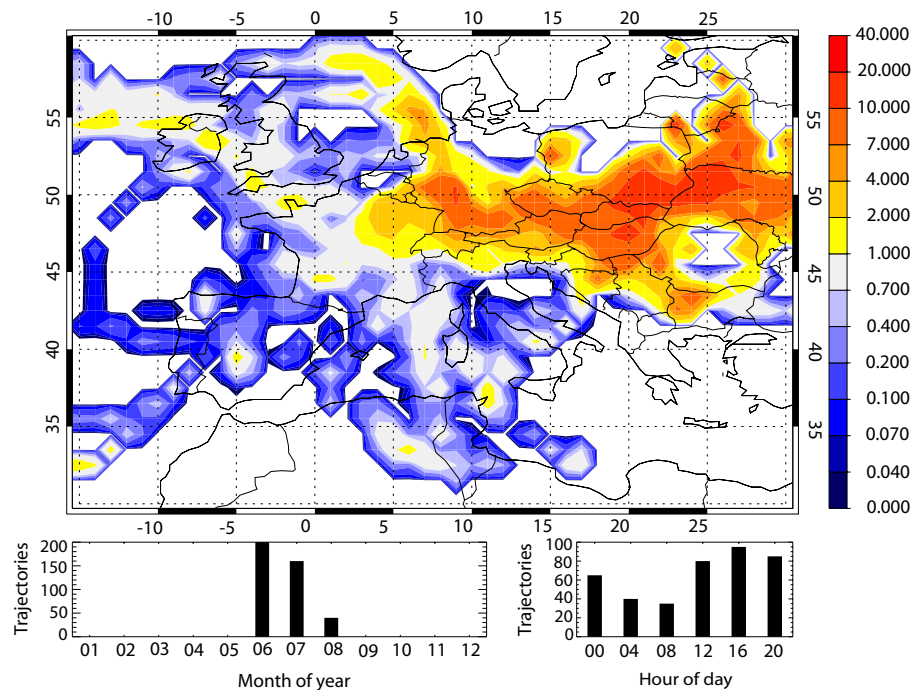


Fig. 12. Relative footprint map associated with low CO_2 and low O_2 and APO concentrations.

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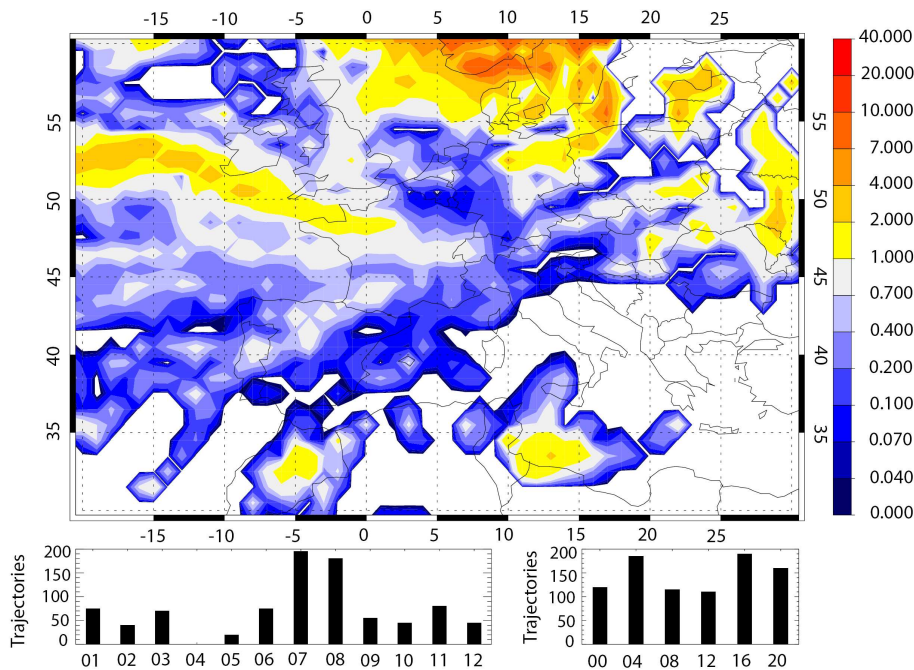


Fig. 13. Relative footprint map corresponding to negative CO_2 but positive APO and O_2 .

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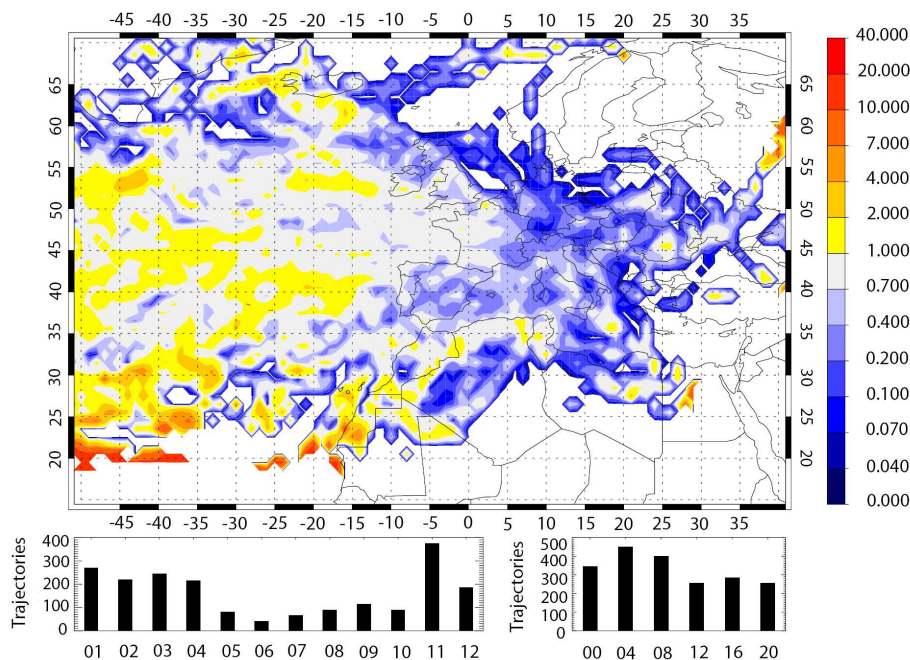


Fig. 14. Relative footprint map associated with high APO (and also high O_2) and low CO_2 . The main provenance of air masses is from the ocean, thus supporting the view of marine biospheric activity being responsible for high APO and oxygen concentrations measured at Jungfrauoch.

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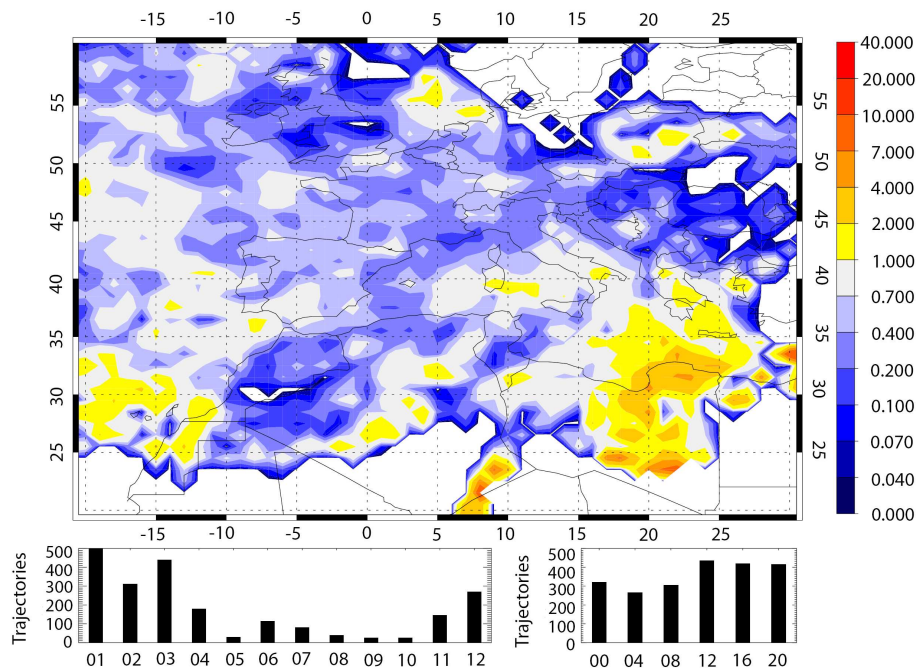


Fig. 15. Relative footprint map associated with regular CO_2 and low O_2 concentrations.

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