Discriminating free troposphere conditions and planetary boundary layer influence at Jungfraujoch

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1. Project description

Empa launched its first atmospheric measurements at Jungfraujoch in 1973 as part of an early engagement of Switzerland in a programme initiated by the Organisation for Economic Co-operation and Development (OECD). In 1978, Empa and the Swiss Federal Office for the Environment (FOEN/BAFU) established the Swiss National Air Pollution Monitoring Network (NABEL), with Jungfraujoch (JFJ) being one of the first 8 sites. In 1990/1991, the NABEL network was extended to 16 monitoring stations that are distributed across Switzerland. The monitoring stations represent the most important air pollution levels ranging from the urban kerbside to the remote background.

Empa's current measurement program at Jungfraujoch includes continuous in-situ analyses of air pollutants such as ozone (O_3), carbon monoxide (CO), nitrogen monoxide (NO), nitrogen dioxide (NO_2), sulphur dioxide (SO_2), and particulate matter. The latter are measured as PM10 (particles < 10 µm), PM2.5 (< 2.5 µm), PM1 (< 1 µm), and the particle number concentration (PNC) between 0.18 and 18 µm. In addition, the greenhouse gases carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) are monitored. All these data are stored as 10-min averages. An extended set of halocarbons (e.g. CFCs, HFCs), sulphur hexafluoride (SF_6) and a selection of volatile organic compounds (alkanes, aromatics) are measured with a time resolution of two hours. Daily samples are taken to quantify particulate sulphur and PM10.

2. Identification of free tropospheric conditions

Due to its altitude, Jungfraujoch is predominantly exposed to pristine air masses representing the lower free troposphere over Central Europe. However, as the station is surrounded by highly industrialized regions at much lower altitudes, influence of (less pristine) air masses from lower atmospheric layers to Jungfraujoch is occasionally observed, driven for example by synoptic phenomena like frontal passages or by thermally induced flow systems during fair weather days. An analysis of station representativeness for a variety of European atmospheric monitoring stations categorized Jungfraujoch as "mostly remote" (Henne et al., 2010). A more recent study analysing the local topography around 43 high altitude stations world-wide identified Jungfraujoch as one of the three stations with lowest boundary layer influence in Europe (Collaud Coen et al., 2018).

Separating the observed time series at Jungfraujoch into subsets according to its exposure can be of interest for various reasons: (a) data sampled under free tropospheric conditions are characterized by an enlarged spatial representativeness giving the subset an eminent status, especially when investigating large scale phenomena and processes in the background atmosphere; the selection of free tropospheric data also allows for a more robust and more representative trend determination as the statistical regression will be less affected by sporadic and temporally irregular planetary boundary layer (PBL) influences; (b) the remainder of the data, i.e. the non-background data, can be used to investigate the underlying transport processes and to quantify the source and sink strengths for specific pollution events and as part of inverse emission modelling; background data can be used to determine the hypothetical baseline conditions during these events, and excess concentrations above the baseline can be determined; (c) transport regimes and processes can be specifically investigated for background and non-background conditions.

The discrimination between free troposphere conditions and episodes under the influence of air masses originating from the planetary boundary layer is a challenging issue. For one, the highly variable and complex structure of the atmospheric boundary layer in complex terrain differs from that over flat terrain by potentially featuring an additional layer that is sporadically mixing with the lower boundary layer ('injection layer', 'aerosol layer'). In addition, the effects of PBL influence differs strongly across various trace gases and aerosol parameters. The degree of this influence depends on the absolute tropospheric burden, the free troposphere to boundary layer gradients, and the reactivity and lifetime of the species.

At mountain stations, a simple restriction of the in-situ data to nighttime conditions can be an easy and straightforward approach that eliminates most of the events with local to regional influence during fair-weather conditions, but may not suffice in the presence of persistent residual layers. Such a basic filtering is for example applied for data compilations of a large number of stations, like the Obspack data release (Schuldt et al., 2021) because a station-tailored but comparable sophisticated selection method (as presented below) is beyond the scope of the Obspack collection activity. At Jungfraujoch, the application of approaches to discriminate free troposphere conditions and planetary boundary layer episodes has a long history. Below, a summary of selected publications is given that either qualitatively describe the PBL influence at Jungfraujoch or quantitatively provide estimates of the frequency of such events.

Filtering based on synoptic patterns

First efforts focussed on a combination of trace gas observations at different altitudes across Switzerland, meteorological data, early versions of transport modelling, and a coarse parametrization of synoptic patterns by the Alpine Weather Statistics (Forrer et al., 2000). The meteorological filtering was refined later to identify periods subject to potential perturbation due to local or regional pollution sources (Zellweger et al., 2003). In this publication, free tropospheric conditions were separated from Foehn events, thermally induced convection and synoptic lifting to the Jungfraujoch station. Figure 1 illustrates the frequency of the occurrence, separated for autumn/winter and spring/summer periods. Free tropospheric conditions are less frequent in spring/summer than in autumn/winter mainly due to the stronger appearance of thermal convection when solar irradiation is higher.

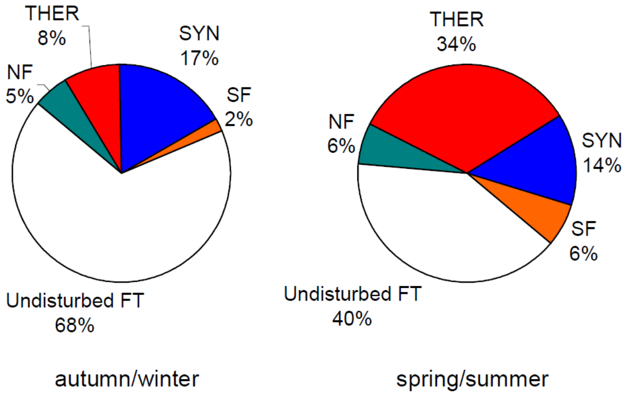

Figure 1. Frequency of meteorological conditions at Jungfraujoch for the period between April 1997 and March 1999 (NF = north foehn, SF = south foehn, THER = thermally induced vertical transport, SYN = synoptic lifting). Autumn/winter refers to September to February, spring/summer to March to August (from Zellweger et al. (2003), in accordance with creative commons licence CC BY-NC-SA 2.5).

Collaud Coen et al. (2011) applied the Alpine Weather Statistics to an extended set (fourteen years) of meteorological parameters, aerosol variables, and trace gases. They showed that increases in carbon monoxide (CO) concentrations due to export of PBL air masses to Jungfraujoch were most pronounced during convective anticyclonic weather conditions.

Water vapour as a proxy

The usefulness of atmospheric water vapour as a tracer for investigating the vertical transport of air masses from the PBL into higher atmospheric layers has been shown by Henne et al. (2005). Cooper et al. (2020) also used the specific humidity as proxy for PBL influence at mountain stations. They compared the humidity at Jungfraujoch with the humidity measured on-board aircrafts within the In-Service Aircraft for the Global Observing System (IAGOS) program at 650 hPa above Western Europe. Due to generally larger specific humidity values at Jungfraujoch, especially in summer during daytime, they concluded that Jungfraujoch observations are not fully representative of the lower free-troposphere and that data need to be carefully filtered to avoid boundary layer air contributions.

Filtering based on trace gas – trace gas ratios

The Free Tropospheric Experiment (FREETEX) campaign in 1998 aimed at improving the understanding the ozone production efficiency in the free troposphere (Carpenter et al., 2000). As part of the campaign, additional observations were implemented to record key parameters being important for ozone photochemistry. One of the newly measured parameters was NOy, which represents the sum of oxidized nitrogen species. Carpenter et al. (2000) used criteria of CO below 200 ppb and a NOx to NOy ratio of smaller than 0.3, with NOx being the sum of nitrogen monoxide (NO) and nitrogen dioxide (NO_2). Shortly after, this approach was modified by Zellweger et al. (2003), who used the NOy to CO ratio as an indicator for rather fresh emissions. Due to their different lifetimes in the atmosphere, NOy to CO ratios were found to be a useful proxy already in the early 1990s. Jaeglé et al. (1998) have shown over the United States that NOy/CO ratios were on the order of 0.1 close to anthropogenic sources, while values of 0.005 were observed in the upper troposphere. Since the publication of Zellweger et al. (2003), the NOy/CO ratio at Jungfraujoch was used by several studies (e.g. Zanis et al., 2007; Pandey Deolal et al., 2013, Herrmann et al., 2015; Lacher et al., 2018) to identify free tropospheric conditions at Jungfraujoch. NOy/CO thresholds used, depending on the study and season, were between 0.03 and 0.08, with values below the threshold being selected as representative for the free troposphere. Due to trends in emissions and concentrations in the boundary layer for both carbon monoxide and the nitrogen species, threshold values likely need to be adjusted over time when applying to long-term observations.

Filtering based on Radon concentrations

Radon is a radioactive noble gas, which is naturally emitted from land surfaces. Radon-222 (^{222}Rn) is the most stable isotope of radon. Radioactive decay is its only sink in the atmosphere, resulting in an atmospheric half-life of 3.8 d. This makes ^{222}Rn a useful tracer of surface influence. Several studies have shown the value of Radon measurements at Jungfraujoch: Griffiths et al. (2014) analysed diurnal ^{222}Rn cycles at Jungfraujoch and Bern for 2010 and 2011 and estimated that boundary layer air masses reached the Jungfraujoch station on about 40% of the days from April to September. Chambers et al. (2016) compared ^{222}Rn concentrations at Jungfraujoch, Mauna Loa (a remote high-altitude island site on Hawaii), and Cape Grim (a coastal low-altitude site in Tasmania). The identification of the least terrestrially influenced air masses revealed that the definition of a generic ^{222}Rn threshold selecting background air masses is unfeasible because Jungfraujoch, located in Central Europe, never reaches ^{222}Rn values that are as low as at Mauna Loa or at Cape Grim. Griffiths et al. (2014) showed that a ^{222}Rn threshold of 1000 mBq m^{-3} was suited to identify aerosol scattering coefficients representative for background. Chambers et al. (2016) concluded that concentrations of selected trace gases and water vapour remain particularly low for ^{222}Rn below 300 mBq m^{-3}. Most recently, Brunner et al. (2021) determined the probability of Jungfraujoch's exposure to free tropospheric conditions by fitting two lognormal distributions to the observed bimodal frequency distribution of 11 years of ^{222}Rn data at Jungfraujoch. The distribution representing free tropospheric conditions showed a mean value of about 726 mBq m^{-3}, while the other distribution, corresponding to conditions under PBL influence, was centred around 2529 mBq m^{-3}.

Statistical filtering

The atmospheric composition in well-mixed free tropospheric air masses is usually characterized by rather low short-term variability. This allows to apply statistical methods to the observed time series for background condition identification. Unlike the other methods mentioned above, statistical approaches usually do not require additional observations next to the target species. This makes them rather universally applicable. However, various parameters of the statistical filtering can be usually changed (e.g. fitting windows, thresholds, exclusion criteria, etc.) and need to be adapted according to the characteristic of the target species and the specific conditions of a station. An early approach for analyzing and filtering CO_2 data from Mauna Loa was published already in 1989 (Thoning et al., 1989). Later, other applications of statistical methods were developed and applied to Jungfraujoch data, like the Robust Extraction of Baseline Signal (REBS) (Ruckstuhl et al., 2012) or the Adaptive Diurnal minimum Variation Selection (ADVS) (Yuan et al., 2018).

Filtering according to atmospheric transport modelling

Atmospheric transport modelling, and especially its application to calculate backward trajectories to investigate the origin of the air masses arriving at Jungfraujoch, became a common approach in recent years. Improvements of the models, better temporal and spatial resolution, and more powerful computational resources allow reliable estimates of the movement of the air masses prior to their arrival at Jungfraujoch. Applications of several models to a variety of Jungfraujoch time series have been published in the last 15 years (Reimann et al, 2008; Cui et al, 2011; Keller et al., 2011; Herrmann et al. 2015; Yu et al., 2020). Figure 2 shows results from a 20-day backward trajectory analysis for the 1990 to 2008 period. This analysis identified about 15-20% and 20-25% of the air masses arriving at Jungfraujoch to be of free tropospheric origin in spring and summer, respectively.

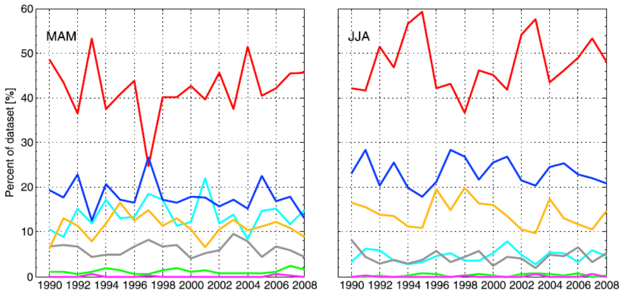

Figure 2. Frequency of the origin of the air masses arriving at Jungfraujoch in spring (left) and summer (right): European PBL (red); free troposphere (blue); North American PBL (yellow); marine boundary layer (sky blue); stratosphere (dark grey); African PBL (magenta); and Asian PBL (green) (from Cui et al. (2011), with permission).

Herrmann et al. (2015) compared in their analysis three different methods to select free tropospheric conditions at Jungfraujoch. The results are shown in Figure 3. The three methods – ^{222}Rn, NOy/CO, and contact of the backward trajectories with the planetary boundary layer – confirmed previous findings in terms of seasonal variations. Free tropospheric conditions at Jungfraujoch were more prevalent in fall and winter than in spring and summer. However, the three methods did not fully agree in terms of the occurrences for the individual months.

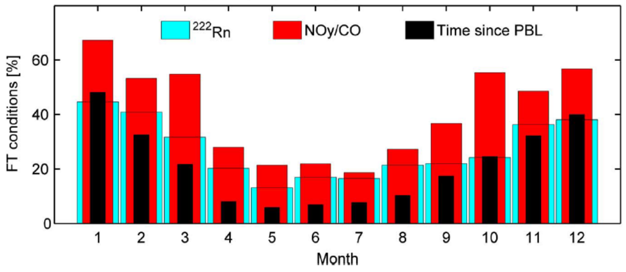

Figure 3. Relative occurrence of free tropospheric (FT) conditions at Jungfraujoch (from Herrmann et al. (2015), with permission).

Most recently, Pieber et al. (2021) combined results from global and European model simulations and emissions inventories. They determined background concentrations by the global model at the boundaries of the regional, European, model.

Boundary layer height determination with ceilometer

In August 2014, a ceilometer was installed at the Kleine Scheidegg, about 4.5 km north-west and 1.5 km below Jungfraujoch. The backscatter signal of the ceilometer allows the determination of the height of the convective boundary layer height and the continuous aerosol layer above the instrument, both proxies for the height of the planetary boundary and aerosol layer around Jungfraujoch (Poltera et al.; 2017). Standardized operational retrieval algorithms are currently developed (Kotthaus et al., 2020). Planetary boundary and aerosol layer height data retrieved from the ceilometer data were recently used in a study by Affolter et al. (2021).

Conclusions

The identification and selection of most-representative conditions with least local to regional influence is a common issue, especially for background monitoring stations at high altitudes or at the coast. For coastal station with clearly defined sectors for advection of pristine marine air masses, the local wind direction can be a good parameter to separate clean air conditions from episodes with local to regional influence. This is, however, an unsuitable approach for Jungfraujoch due to the location in Central Europe and the topography surrounding the station that strongly affects the local wind measurements. As shown in this review, many different approaches were developed over the last 25 years. However, no clear best-performing method can be recommended since a station-specific one-fits-all approach does not exist. The method of choice needs to be selected (and likely adapted) according to the species of interest and the research question. Similarly, the application of a common approach to several monitoring stations is also often inappropriate. Different station characteristic such as altitude, topography, climatology, exposure, vicinity to different sources etc. usually require an adjustment of the selection procedure.

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https://icos-atc.lsce.ipsl.fr/JFJ

Collaborating partners / networks

Bundesamt für Umwelt (BAFU) / Federal Office for the Environment (FOEN)
Belgian Institute for Space Aeronomy, Brussels
Climate and Environmental Physics, University of Bern
Environmental Geosciences, University of Basel
Institut d'Astrophysique et de Géophysique, Université de Liège
Institute for Atmospheric and Climate Science, ETH Zurich
Laboratory of Atmospheric Chemistry, Paul Scherrer Institut
MeteoSchweiz
World Meteorological Organisation (WMO)
ACTRIS – Aerosol, Clouds, and Trace Gases Research Network
EMEP – European Monitoring and Evaluation Programme
GAW – Global Atmosphere Watch
ICOS – Integrated Carbon Obervation System Research Infrastructure
IG3IS – Integrated Global Greenhouse Gas Information System
NABEL – Swiss National Air Pollution Monitoring Network
VAO – Virtual Alpine Observatory

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