

Quality assurance and quality control of CO₂ observations

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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 organised by the Organisation for Economic Co-operation and Development (OECD). In 1978, Empa and the Swiss Federal Office for the Environment (FOEN/BAFU) initiated 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. The NABEL site at Jungfraujoch is such a remote site; it represents the lower free troposphere in central Europe.

The current measurement program at Jungfraujoch includes continuous in-situ analyses of ozone (O₃), carbon monoxide (CO), nitrogen monoxide (NO), nitrogen dioxide (NO₂), sulphur dioxide (SO₂), methane (CH₄), carbon dioxide (CO₂) and nitrous oxide (N₂O). Particulate matter is measured as PM₁₀ (particles < 10 µm), PM_{2.5} (< 2.5 µm), PM₁ (< 1 µm), and the particle number concentration (PNC) between 0.18 and 18 µm is also recorded continuously. All these data are stored as 10-min averages. An extended set of halocarbons (e.g. CFCs, HFCs), sulphur hexafluoride (SF₆) and a selection of volatile organic compounds (VOCs) (alkanes, aromatics) are measured with a time resolution of two hours. Daily samples are taken to quantify particulate sulphur and PM₁₀.

A comprehensive calibration, quality assurance (QA) and quality control (QC) strategy is of utmost importance when performing atmospheric composition measurements. This is particularly true when measurements are long-term and part of a network of stations to ensure that the observed variability and trends are not due to changes in the instruments' sensitivity, and to allow for a best possible compatibility of the different time series within the network or across several networks, respectively. The latter also requires a well-documented traceability to internationally agreed scales, which are maintained by either National Metrology Institutes or assigned Central Calibration Laboratories in case of the Global Atmosphere Watch (GAW) programme of the World

Meteorological Organisation (WMO). The required quality and quantity of the data are defined by so-called data quality objectives (DQOs). DQOs specify tolerable levels of measurement uncertainty and/or network compatibility based on the scientific question of interest or the decisions to be made (WMO, 2017). Within the context of GAW, DQOs are released as part of GAW reports (available via the WMO library, <https://library.wmo.int/>) or in the peer-reviewed literature as those publications often have a wider visibility in the scientific community. Sometimes, several resources are available, for example in the case of tropospheric ozone. There, DQOs for traditional GAW goals such as the detection of long-term changes in ozone background concentrations are given in a GAW report (WMO, 2013), but more explicit requirements for specific scientific questions, such as requirements for the validation of air quality models or satellite-borne ozone data, were released in a later publication (Tarasick et al., 2019).

DQOs are particularly demanding for carbon dioxide (CO₂) observations due to its long atmospheric lifetime, the strong interaction with the biosphere and consequently only small spatial gradients. The DQOs for greenhouse gases like CO₂ are regularly reviewed during the bi-annual Meeting on Carbon Dioxide, Other Greenhouse Gases and Related Measurement Techniques – GGMT (WMO, 2020). The desired compatibility goal, understood as "*the scientifically-determined maximum bias among monitoring programmes that can be included without significantly influencing fluxes inferred from observations with models*" for measurements in well-mixed background conditions is 0.1 ppm in the Northern hemisphere. To meet this goal, an internal reproducibility including instrumental imprecision, uncertainties in the traceability chain or uncertainties due to gas handling and drying of about 0.05 ppm (or roughly 0.1 permil at a level of 400 ppm) needs to be achieved (WMO, 2020). These specifications require high-precision instrumentation, a comprehensive QA/QC strategy, and a close and unbroken link to one of the internationally accepted primary scales. Operators of European greenhouse gas observations in pristine regions teamed up under the umbrella of the Integrated Carbon Observation System (ICOS) Research Infrastructure (www.icos-ri.eu). Members of ICOS, which is recognized as a contributing network to GAW, benefit from access to a Central Analytical

Laboratory in Germany, which provides reference gases directly traceable to the WMO scales maintained by GAW's Central Calibration Laboratories. For an optimum compatibility, ICOS also developed detailed specifications in terms of operation, like number of calibration gases, and specifies requirements in terms of sample periphery, like drying units, tubing material, models of pressure regulators etc. (ICOS RI, 2020). ICOS stations also undergo a rigorous assessment prior to acceptance to the network. The assessment procedure is documented in a recent publication by Yver-Kwok et al. (2021). A minimum of three high-pressure cylinders are required for a weekly to bi-weekly calibration, and two additional target tanks are used to track the short-term and longer-term performance of the instrumentation. Data from all ICOS stations are sent daily to the Atmospheric Thematic Centre (ATC) in France for centralized data processing. There, the data sets are split into subsets of in-situ (ambient air) data, target data and calibration data and further processed (see Figure 1 for illustration). Target data are measured up to once per day, are treated as unknowns in the same way as the in-situ data and are used as quality indicators and timely markers in case of problems.

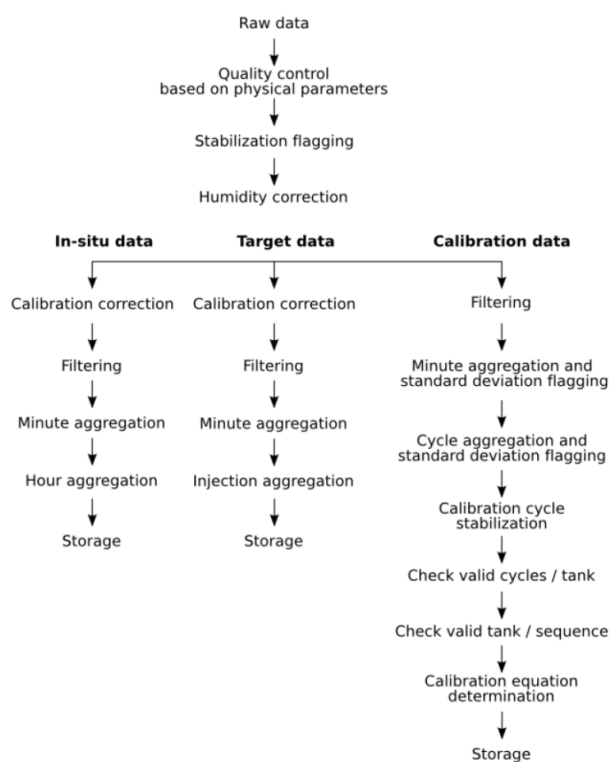


Figure 1. Schematic of data treatment during automatic processing at the ICOS Atmospheric Thematic Centre (taken from Hazan et al., 2016).

Standardized output is generated for visual inspection and quality control and is made publically available on the ICOS ATC panel board (<https://icos-atc.lscce.ipsl.fr/dp>). One of those plots is shown in Figure 2. The multi-panel figure shows the ambient air data on top, followed by the standard deviation of the ambient air data, the continuous measurement repeatability (CMR) defined as the monthly average of the standard deviations of short-term target raw data over 1 min intervals, the long-term repeatability (LTR) calculated as the standard deviation of the averaged short-term target measurement intervals over 3 days, and the difference of the calibrated short-term target data to its assigned value. The enhanced levels of LTR uncertainty and short-term target bias at the beginning of the period were caused by a problem with the

sample drying. These numbers could be flattened when changing the drying unit in early November 2017. A series of other automatically generated plots are updated once a day and are made available on the ATC webpage.

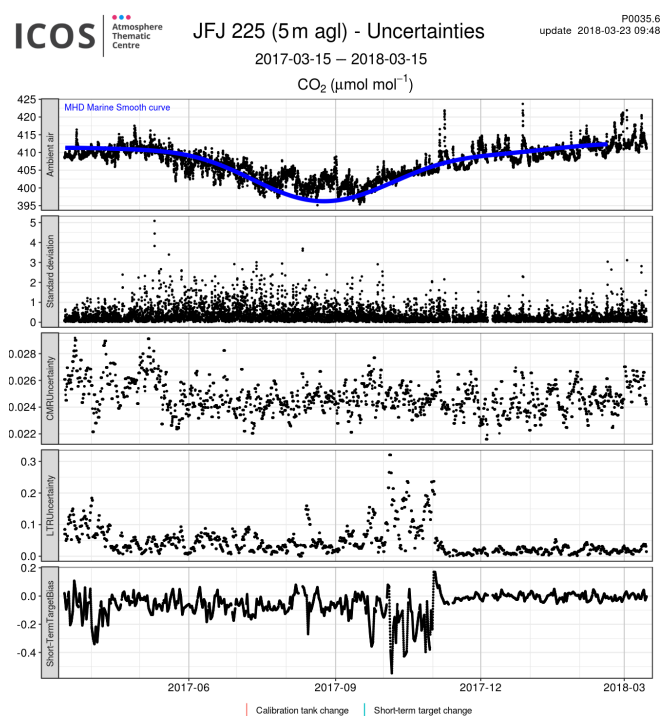


Figure 2. Standardized ICOS summary plot for atmospheric CO₂ data from Jungfraujoch. See the text for details. The smoothed curve for Mace Head (in blue) is added for visual guidance to the plots of all stations to simplify the comparison (taken from Yver-Kwok et al., 2021).

Next to the background station Jungfraujoch, CO₂ is also measured at the NABEL stations Haerkingen, Payerne and Rigi-Seebodenalp. The observations close to the highway in Haerkingen started in 2007 mainly as an indicator for the intensity of the traffic influencing the site. In more recent years, CO₂ measurements were also launched at Payerne and Rigi-Seebodenalp, partly facilitated through the availability of multi-species analyzers allowing to measure CO and CO₂ with the same instrument. In 2019, calibration and traceability of the CO₂ measurements at Payerne and Rigi-Seebodenalp were also implemented. Due to the rather large variability at Haerkingen, Payerne and Rigi-Seebodenalp (see Figure 3), a relaxed QA/QC strategy can be followed, and a simplified calibration scheme is applied. Instruments are regularly calibrated with CO₂-free air and one reference tank. Concentrations in the reference tanks are assigned by WMO/GAW's World Calibration Centre for CO₂ at Empa and are, thus, also traceable to the common GAW scale. The atmospheric variability on the Swiss plateau (observed at Haerkingen and Payerne) is largely driven by meteorology, such as the intensity of venting of the plateau or the occurrence of inversions. The measured CO₂ concentrations at Haerkingen are usually higher than at Payerne due to the vicinity of the highway, but persistent episodes with particularly low (e.g. in late February/ early March 2020) or high concentrations (e.g. in mid-March 2020) can be observed at both sites. The elevation of the Rigi-Seebodenalp station (1031 m asl) results in significant less variability than on the Swiss plateau but there is still more scatter than at Jungfraujoch, which is mostly exposed to free tropospheric air masses.

The CO₂ measurements at the four NABEL stations also serve as reference for the recently established CarboSense network (<http://carbosenet.wikidot.com>). The CarboSense network is a uniquely dense CO₂ sensor network across Switzerland, which includes more than 250 low-cost sensors measuring CO₂. Due to the large number of sensors, network control and sensor calibration requires novel QA/QC approaches. Prior to deployment, all sensors are calibrated in a pressure and climate chamber as well as in

ambient conditions when being exposed next to a reference instrument for several weeks. Once deployed, sensor data are corrected for interferences of atmospheric water vapour, followed by an outlier and drift correction and a final consistency check (Müller et al., 2020). The high-precision NABEL data are used for sensor calibration and assessment of the sensors' long-term performance as well as for correcting the sensor drifts during field operation.

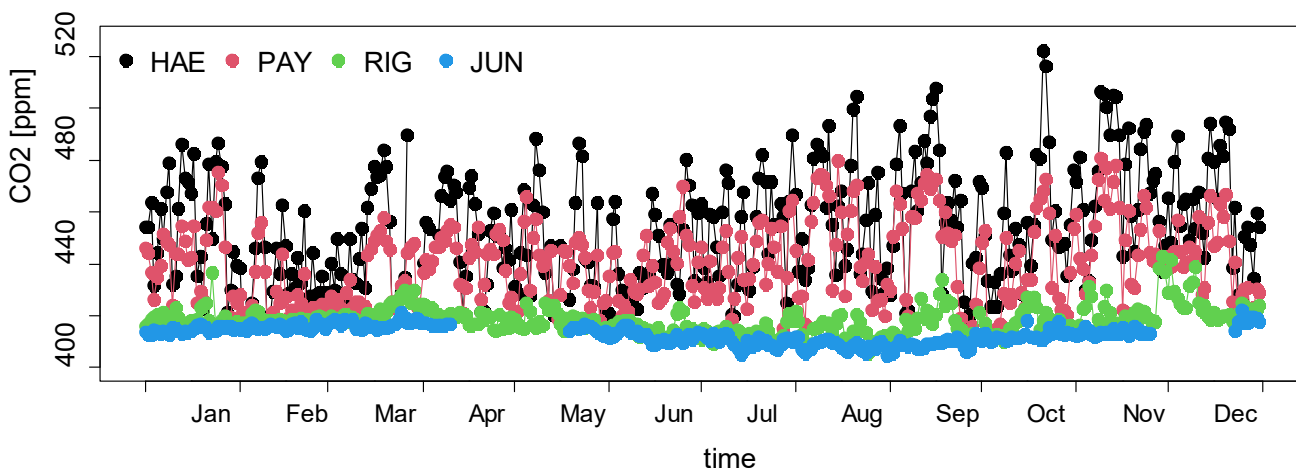


Figure 3. Daily averages of CO₂ mole fractions in 2020 at Jungfrauoch (blue) and other lower elevation sites of the Swiss National Air Pollution Monitoring Network, Haerkingen (HAE, black), Payerne (PAY, red), and Rigi-Seebodenalp (RIG, green).

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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 for 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 Observation System Research Infrastructure
 IG3IS – Integrated Global Greenhouse Gas Information System
 NABEL – Swiss National Air Pollution Monitoring Network
 RINGO – Readiness of ICOS for Necessities of Integrated Global Observations

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