Flux calculation using CARIBIC DOAS aircraft measurements – SO_2 emission of Norilsk

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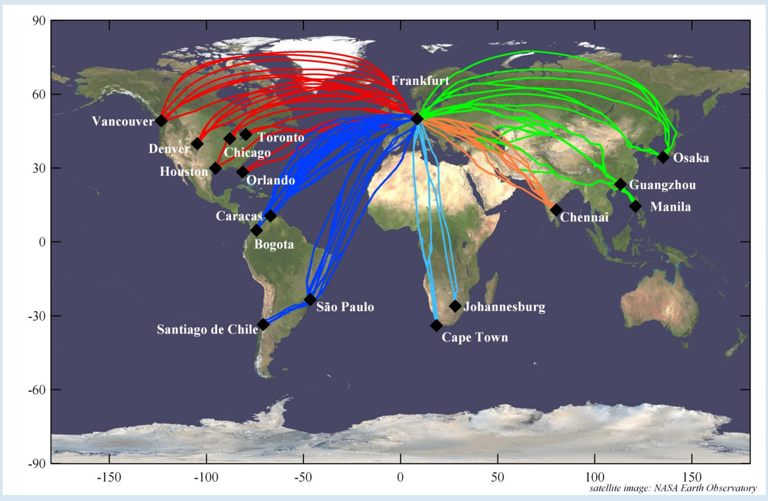
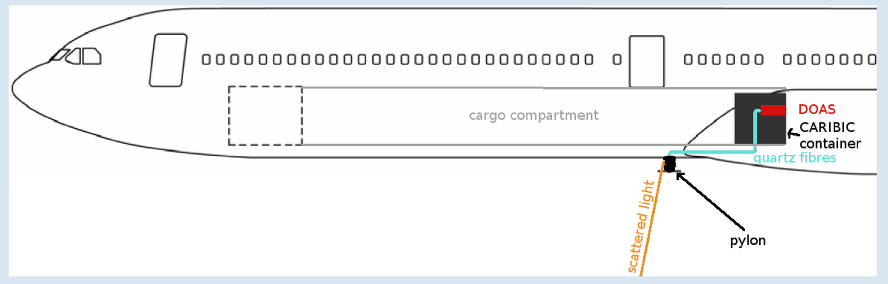
THE CARIBIC PROJECT

Civil **A**ircraft for the **R**egular **I**nvestigation of the atmosphere **B**ased on an **I**nstrument **C**ontainer

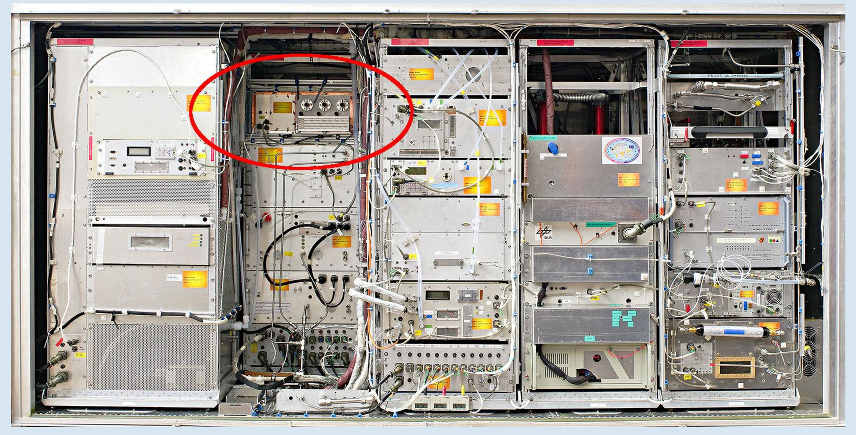


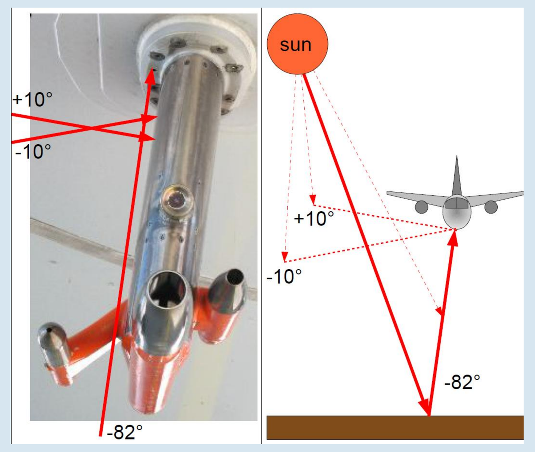
CARIBIC is a long-term airborne measurement system for observing the atmosphere during regular passenger flights. The aim of CARIBIC is to provide detailed data for a better understanding of atmospheric processes like long range transport of pollution or stratosphere-troposphere exchange.

The inlet system (CARIBIC pylon) is installed permanently under the aircraft's fuselage. It has three inlet tubes, one for water, one for aerosols and one for trace gases. The CARIBIC pylon also houses three DOAS telescopes.



Once a month the fully automated measurement container (weight: 1.6 tons) is installed in the cargo compartment for four consecutive flights. It contains twelve in situ instruments (trace gases, aerosols, humidity), and 116 air sample flasks. The DOAS system for remote sensing forms the third category of equipment.





THE DOAS INSTRUMENT

The three small telescopes for the DOAS system in the inlet system receive light from three directions (10° above horizon and 10° and 82° downwards). Quartz fibres conduct the light to three spectrographs (wavelength range 286–423 nm) inside the container.

While the nadir telescope (-82°) gets light being reflected from the surface as well as light scattered in the air between the aircraft and the surface, the other telescopes (+10° and -10°) typically are more sensitive for altitudes above and slightly below the aircraft. The instrument setup is optimized for NO_2 and SO_2, but also BrO, HONO, HCHO, O_3, O_4, and OClO are included in the DOAS retrieval.

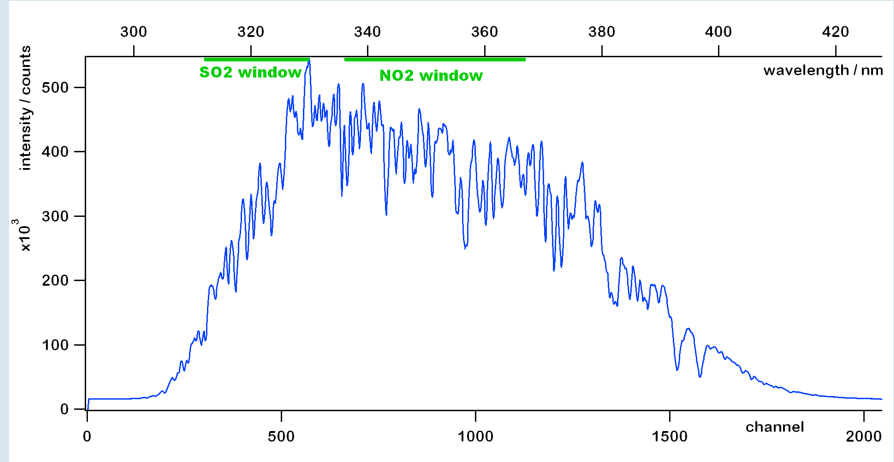
Differential Optical Absorption Spectroscopy (DOAS) is based on the Lambert-Beer's law.

$$I(\lambda) = I_0(\lambda) \cdot e^{-\left(\sum_i \sigma_i(\lambda) \cdot S_i + \varepsilon_{scat}(\lambda) \cdot L\right)}$$

When light passes through an absorbing layer like the atmosphere, the initial intensity $I_0(\lambda)$ is attenuated due to absorption and scattering to the intensity $I(\lambda)$ at the instrument. Literature cross sections σ_i of several gases can be fitted simultaneously, obtaining the so-called Slant Column Densities (SCD) S_i which are the integral of the concentration along the light path:

$$S_i = \int c_i \cdot dl$$

The broad band structured absorbing and scattering ε_{scat} can be approximated by a polynomial.



Typical spectrum recorded by the nadir instrument.

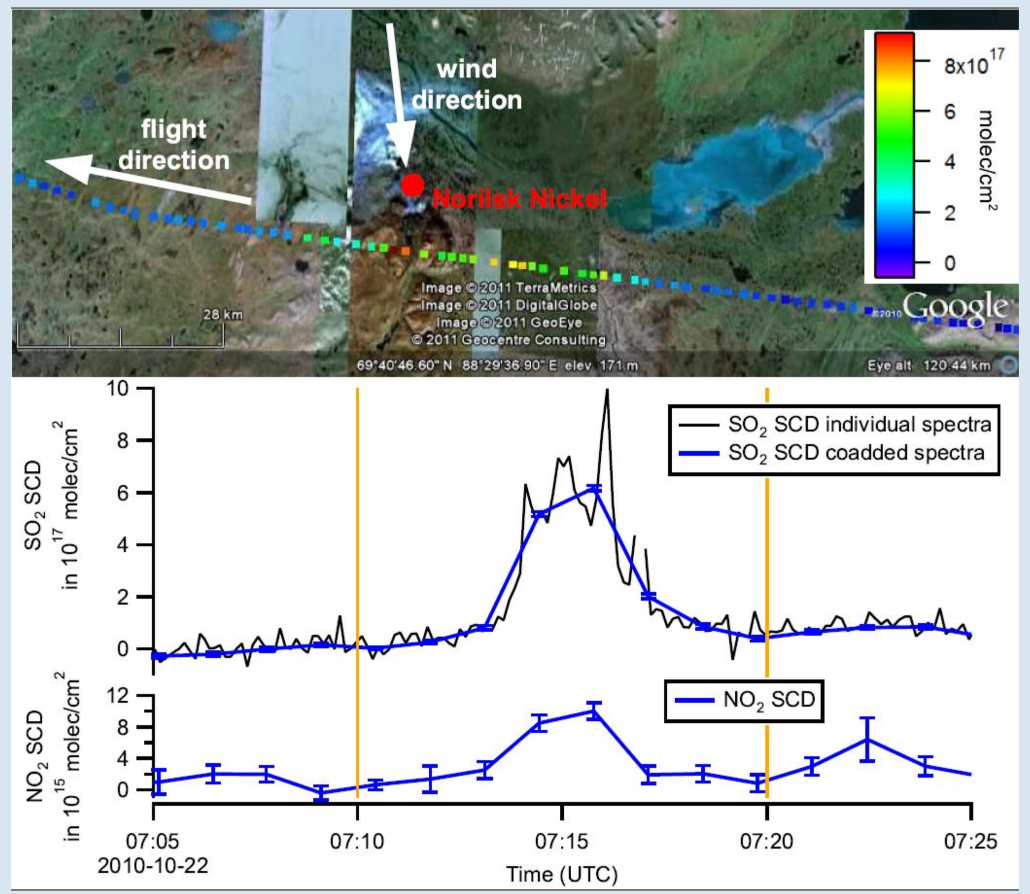
The bottom axis shows the 2048 channels of the CCD while the top axis shows the corresponding wavelength. The green bars indicate the wavelength range used for the retrieval of SO_2 and NO_2 respectively.

NORILSK NICKEL

Norilsk is a city in northern Siberia (69.32°N 88.27°E) with about 130 000 inhabitants.

The principal employer in the area, MMC Norilsk Nickel, is a mining and smelter company, emitting huge amounts of gases and aerosols, making Norilsk one of the most polluted areas in the world.

Beside metallic compounds, several trace gases are emitted, amongst them NO_2 and SO_2.



The SO_2 emitted from Norilsk can be observed by satellites regularly.

On 22 Oct. 2010, the CARIBIC aircraft flew (at 10.6 km altitude) over this region, about 6 km to the south of the industrial plant. As the wind was coming from the north, the SO_2 inside the exhaust plume was observed by the nadir instrument (07:15 UTC).

For NO_2, a second peak was observed at 07:23 when flying over "Dudinka", the seaport of Norilsk Nickel.

Top: Section of the flight route between 07:10 and 07:20 UTC on 22 Oct. 2010, colour-coded with the SO_2 SCD (individual spectra). The red dot shows the position of industrial facilities of Norilsk Nickel.

Bottom: Time series of SO_2 and NO_2. The black line corresponds to the dots in the top part. For reducing noise, co-added spectra were evaluated (blue lines).

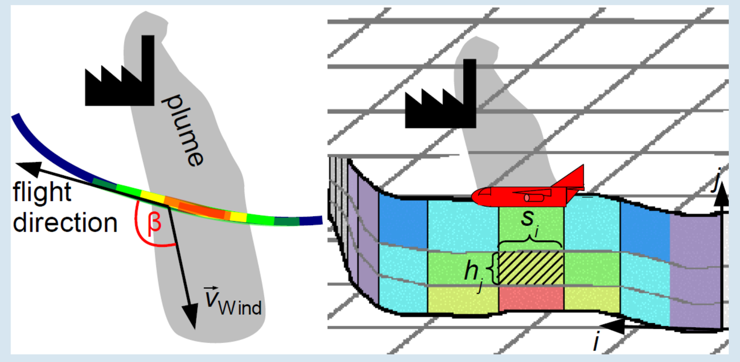
FLUX CALCULATION

The strength of a source (here Norilsk Nickel) can be calculated integrating the flux through an area:

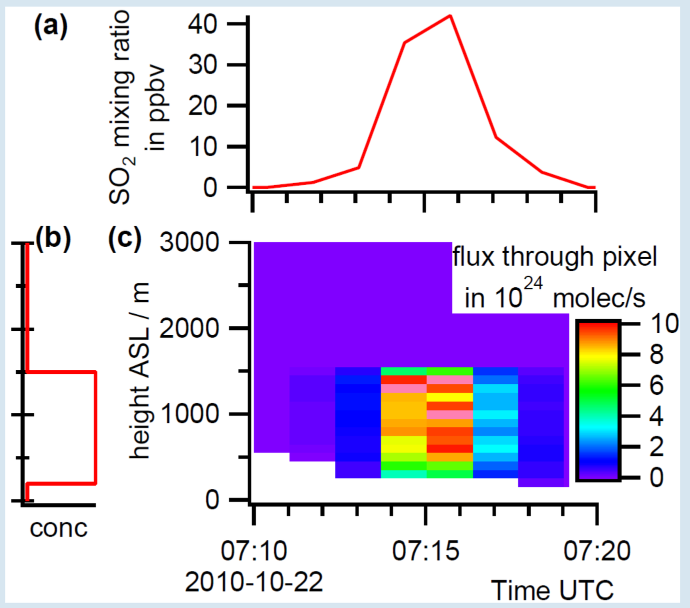
$$Q = \int d\vec{A} \cdot \vec{J} = \int d\vec{A} \cdot c \cdot \vec{v} = \sum_i s_i \cdot \sum_j h_j \cdot c_{i,j} \cdot v_{i,j} \cdot \sin \beta_{i,j}$$

Q: source strength; c: number concentration; A = h·s; J: flux; v: wind speed

Assumed is that the source strength was constant. Also assumed is a constant wind speed, however not necessarily uniform throughout the surface. Furthermore it is expected that the complete plume was observed. Moreover, the plume is assumed to have been well mixed within a mixing layer of about 1.5 km height.



With these assumptions and using wind data from Flexpart simulations (based on ECMWF), the SO_2 flux and therewith the source strength is estimated to have been **$2.8 \cdot 10^{26}$ molec/second** which corresponds to **0.9 Mt/yr** of SO_2.



UNCERTAINTY FACTORS

Beside the extrapolation of our single measurement to one year, the main uncertainty factors for the flux calculation are supposed to be:

- wind field:
  - temporal variations
  - uncertainties in models especially at low altitudes
- surface albedo
  - albedo 0.9 → 0.9 Mt/yr
  - albedo 0.6 → 1.4 Mt/yr (+50%)
- AMF
  - aerosol extinction (0.4/km) →1.5 Mt/yr (+60%)

COMPARISON WITH OTHER MEASUREMENTS

- GOME satellite (Khokhar, Platt, Wagner, 2008): **1.7 ± 0.3 Mt/yr** for 1996 - 2002
- variously reported (according to Carn et al., 2004): **2 - 3 Mt/yr**
- **OMI measurements on the same day**

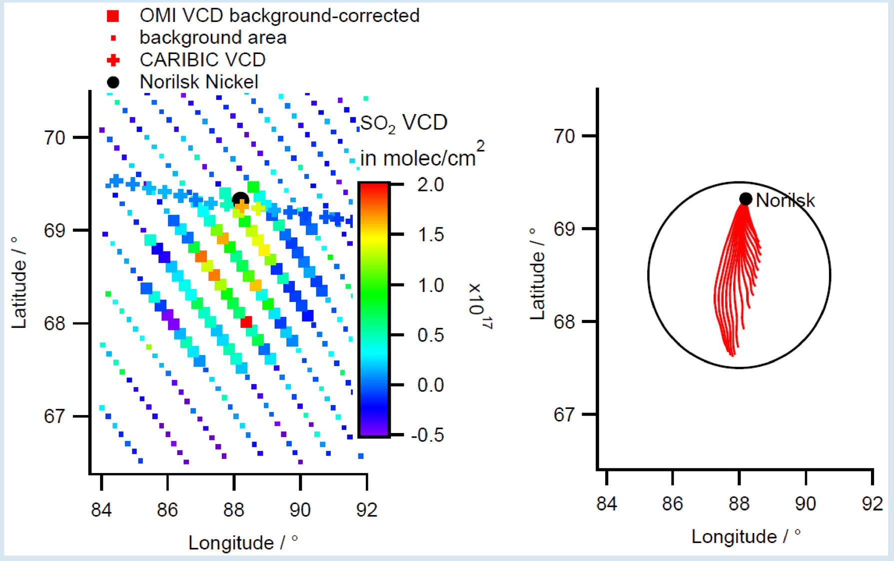
Overpass time: 05:38 UTC, just 1.5 hours before CARIBIC

Assuming Norilsk to be the only SO_2 source in that region and an atmospheric lifetime of SO_2 of τ = 1 day, the daily SO_2 emission can be estimated to have been about $1.8 \cdot 10^{31}$ molecules. This corresponds to about **0.7 Mt/yr**.

This value is obtained by integrating the SO_2 VCD downwind of the Norilsk facilities:

$$E_{annual} = \frac{\int V \cdot dA_{area}}{\tau} \cdot 365\,\text{days}$$

In contrast to the flux calculation, this calculation is independent of the wind speed, but directly antiproportional to the lifetime of SO_2, which is a major uncertainty factor of this approach.



Left: OMI data from the Sulfur Dioxide Space Flight Center, Greenbelt, USA; http://so2.gsfc.nasa.gov/)

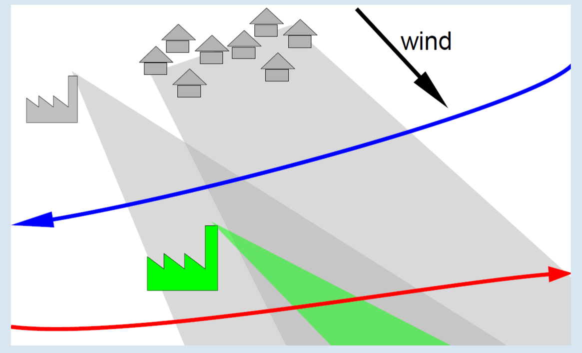
OMI VCD (big squares) after subtracting the average of the surroundig area (small squares). Crosses: CARIBIC flight route.

Right: One-day forward trajectories created through the HYSPLIT web interface.

CONCLUSIONS

- Good agreement between CARIBIC and OMI, using different approaches:
  - OMI: wind not explicitly used, SO_2 lifetime is essential
  - CARIBIC: depends on wind field, SO_2 lifetime of minor significance
- Our result is at the lower end of previously published results, however uncertainties are significant.
- It would be valuable to repeat these measurements.
- Our method also can be applied for other trace gases and other sources, e.g. NO_2 emissions of large cities.
- Further passenger aircraft permanently equipped with DOAS could be combined in a network.

Possible application of an airborne DOAS network: By subtracting the flux under one flight route from the flux under another flight route, it would be possible to distinguish the emission of several sources from one another. If the temporal difference between the flight routes are small enough, this approach is feasible also for sources with a diurnal cycle like cities.



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Please inspect the CARIBIC website **www.caribic.de** *for more information.*