On the detection of African pollution outflow over the Atlantic Ocean using passive DOAS

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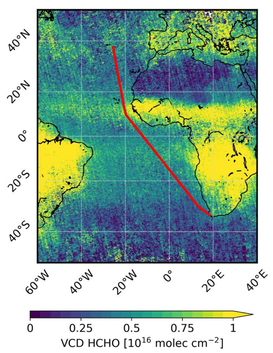
1 Motivation

- enhanced levels of atmospheric pollutants can be identified over the Atlantic Ocean in satellite trace gas maps

⇒ given the very short lifetimes, current knowledge cannot explain their presence far from the coasts → vallidation using independent ground-based measurements is needed



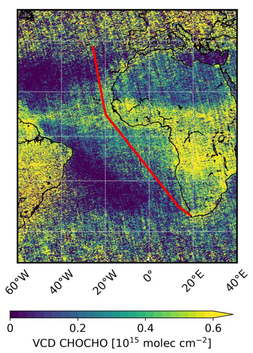


Figure 1: Formalhyde (HCHO) and glyoxal (CHOCHO) satellite vertical column densities (VCDs) for October 2016 measured with OMI
The red line depicts the cruise track of MSM58/2 (see 2 Project, Ship Cruise & Data)

2 Project, Ship Cruise & Data Analysis

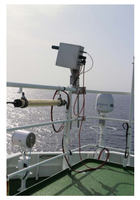


Figure 2: Research vessel (RV) Maria S. Merian
A MAX-DOAS instrument was installed on board for the cruise.

Project:

- COPMAR - Continental Outflow of Pollutants towards the MArine tRoposphere:
- conducted: 8.-25.10.2016; on the RV Maria S. Merian (MSM; Fig. 2)
- cruise: from Ponta Delgada (Azores) to Cape Town (South Africa, Fig. 3)
- the campaign was part of the cruise MSM58/2

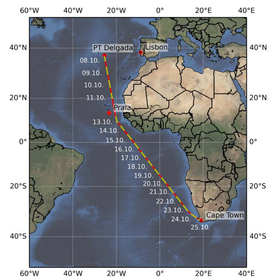


Figure 3: Cruise track
Red: periods of measurement during daylight.
Yellow: the measurement periods (day).

Procedure:

- ship-based Multi-AXis Differential Optical Absorption Spectroscopy (MAX-DOAS) measurements with elevation angles from 0° (horizon) to 90° (zenith) are used for the anaylsis → one scan took ~10 min
- continuous scans of a vertical plane towards the African continent
- data are corrected for the ship's movement (roll)
- measurements potentially contaminated by the vessel plume are excluded (rel. wind direction: 90°-270°)
- only solar zenith angles (SZA) smaller than 70° are used for the analysis of HCHO and CHOCHO, because for lower SZA the measurement uncertainties increases

3 Method

DOAS: Differential Optical Absorption Spectroscopy

- based on Lambert Beer's law: $I(\lambda,s) = I_0(\lambda,s)e^{(-\sigma(\lambda)\rho s)}$
  → the absorption of light travelling through the atmos. can be calculated
- amount of trace gases can be derived from the absorption, resulting in differential slant column densities (dSCDs): $dSCD = \int \rho(s)ds$
  - retrieved with least squares fit
- dSCDs can be converted to VCDs by using differential air mass factors (dAMF), calculated with the radiative transfer model SCIATRAN (Rozanov et al., 2014): VCD = dSCD / dAMF

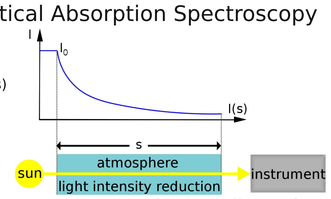


Figure 4: Illustration of the DOAS method
I: reduced intensity
I_0: intensity of the light directly from the sun
s: light path
λ: wavelength
σ: absorption cross-section
ρ: concentration of absorbers

4 Detection of HCHO & CHOCHO

- → pollution close to the ground: largest dSCDs are expected for the lowest elevation angles
- → pollution higher in the atmosphere: largest dSCDs are expected at higher elevation angles
- on three/two days, a different elevation angle dependency can be observed for HCHO/CHOCHO dSCDs (not shown)
  ⇒ pollution higher in the atmosphere
- 13th and 14th October: enhanced HCHO and CHOCHO values are visible in MAX-DOAS, satellite, and model data, regarding MOZART-4 (Fig. 5 & 6)
- 17th October: enhancement only visible in HCHO MAX-DOAS data (Fig. 5)
  - not present in satellite and model data which are monthly means → a single, isolated outflow event on one particular day is not distinguishable from background
- HCHO:
  - MAX-DOAS often observed enhanced columns compared to sat. and model data (Fig. 5)
  - → high uncertainties in sat. data over unpoll. scenes
  - → underestimation of VCDs in sat. and model data: localised nature of the enhancements
- CHOCHO:
  - OMI observations show enhanced columns throughout the tropics (Fig. 6)
  - → satellite data are close to the detection limit of the retrieval
  - MOZART-4 data are mostly close to or slightly higher than our measurements

⇒ African continental outflow of HCHO and CHOCHO can be detected on some days

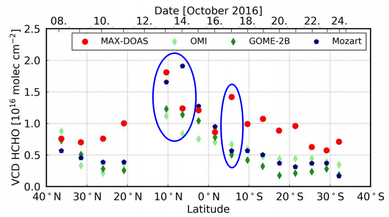


Figure 5: Daily mean HCHO VCDs
Lat. dependency of HCHO VCDs along the cruise track for MAX-DOAS, sat. measurements, and model data. Correlation coefficient of MAX-DOAS & MOZART-4: 0.72; MAX-DOAS & OMI: 0.80; MAX-DOAS & GOME-2B: 0.70.

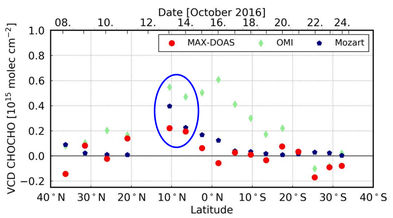


Figure 6: Daily mean CHOCHO VCDs
Same as Figure 7 but for CHOCHO. Correlation coefficient of MAX-DOAS & MOZART-4: 0.55; MAX-DOAS & OMI: 0.56. MAX-DOAS CHOCHO VCDs are close to the detection limit (1.4×10^{14} molec cm^{-2}).

5 Source regions of VOC precursors

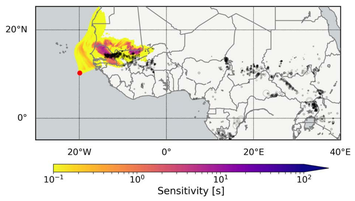


Figure 7: Emission sensitivity on 13.10.
The red dot indicates the mid day position of the RV Maria S. Merian. The emission sensitivities in the lowest 0.5 km for particles arriving at 2.5 km altitude above the ship, integrated over 2 days backward. The black circles are fires which were detected between 10th and 12th October, taken from the FINN database, scaled with the calculated CO_2 emissions.

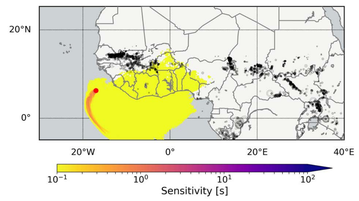


Figure 8: Emission sensitivity on 14.10.
Same as Figure 7, the emission sensitivity is integrated over 4 days backward, in the lowest 1 km. The black circles are fires which were detected between 8th and 13th October.

- emission sensitivities are simulated with FLEXPART
- 13th and 17th October: simulated air tracers originate from the African continent (Fig. 7 & 9)
  - in the source regions are forests, grasslands, and fires
  - altitudes between 1.5–3 km and 2–4.5 km after 2 and 4 days
- 14th October: small sensitivity (Fig. 8) to the continent, the air reaches the ship's position after 4 days in an alt. between 1–5 km

⇒ volatile organic compounds (VOCs) or their precursors have to be transported over long distances from the African continent, contrary to what is expected due to the short lifetime of HCHO and CHOCHO

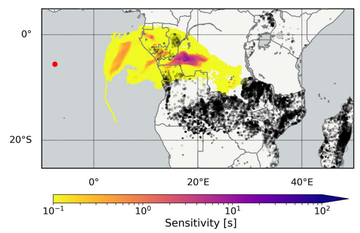


Figure 9: Emission sensitivity on 17.10.
Same as Figure 7, the emission sensitivity is integradted over 4 days backward, in the lowest 0.5 km. The black circles are fires which were detected between 12th and 14th October.

6 Summary

Results from MAX-DOAS measurements

- outflow of HCHO and CHOCHO from the African Continent to the Atlanic Ocean can be observed on three and two days, respectively, in elevated layers
- we found high correlation coefficients between MAX-DOAS measurements and model / satellite data
  - MAX-DOAS HCHO columns are often higher than both satellite and model data, pointing to an underestimation in satellite and model data
  - MAX-DOAS CHOCHO columns are often lower than both satellite and model data, pointing to high uncertainties

Results from the sensitivity study

- on 13th and 17th October: the results suggest that VOCs or their precursors were transported over long distances from the African continent
- on 14th October: the emission sensitivity to the continent is small and high to the open ocean, fires are less important

References & Acknowledgements

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