DOAS RETRIEVAL OF GLYOXAL FROM SPACE

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ABSTRACT

Glyoxal ($C_2H_2O_2$) is formed by the oxidation of several VOCs and hence serves as an indicator for fast VOC chemistry. $C_2H_2O_2$ has characteristic absorption bands in the blue spectral range, allowing remote sensing by DOAS. From space, the detection of enhanced Glyoxal column densities has recently been reported from OMI and SCIAMACHY data.
Here we present the results of our Glyoxal retrieval from SCIAMACHY spectra. In the resulting mean maps, photochemical hot spots due to anthropogenic activities can clearly be identified, for instance Hong Kong or Los Angeles. Also strong biomass burning events lead to enhanced Glyoxal levels. Generally, the detected $C_2H_2O_2$ column densities are high over the tropical rain forest, indicating biogenic emissions. Also over some bioactive oceanic regions enhanced Glyoxal levels are found, possibly indicating VOC emissions over ocean.

1 INTRODUCTION

Glyoxal ($C_2H_2O_2$) is the smallest α-dicarbonyl. It is released by several processes, e.g. biomass burning, fermentation and industrial processes. In addition, $C_2H_2O_2$ is generally formed by the oxidation of (anthropogenic and biogenic) VOCs [1]. Hence Glyoxal serves as indicator for fast VOC chemistry [2].
Glyoxal has characteristic absorption bands in the blue spectral range (see Fig. 1), allowing remote sensing by Differential Optical Absorption Spectroscopy DOAS [3].
Ground based DOAS measurements of Glyoxal have been successfully performed e.g. in Mexico City [2]. From space, $C_2H_2O_2$ detection has recently been reported using data from the OMI satellite instrument [4] and SCIAMACHY [5][6].
The SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY SCIAMACHY on the ESA satellite ENVISAT allows the global retrieval of trace gas column densities of several molecules having absorbing features in the UV/Vis spectral range like e.g. O_3, NO_2, SO_2 or Formaldehyde. Now Glyoxal is a further trace gas being detectable from space, holding valuable information on fast VOC chemistry.

2 RETRIEVAL

The retrieval of slant column densities (SCDs) of Glyoxal using DOAS [3] is analogue to the NO_2 retrieval from SCIAMACHY as described in [7] (fitting O_3, H_2O, NO_2, and O_4 cross sections, a Ring spectrum, and a 3rd degree polynomial), adding the Glyoxal cross section from [8]. For our analysis, we use the spectral range 436.1 - 457.4 nm. Fig. 1 shows an example of the Glyoxal fit (with earthshine reference) for an overpass over southeast Australia during a strong biomass burning event on 18 January 2003.
When using the solar reference measured by SCIAMACHY daily, the fit quality suffers from artificial spectral features due to the diffuser plate, etalon structures, and possibly calibration issues, leading to structured residuals and offsets in the resulting slant column density (SCD). Therefore, the retrieved Glyoxal SCDs are normalized with respect to the Sahara where a zero level is assumed.
Over oceans, the fit does not work well, probably caused by spectral structures of liquid water and/or organic matter (phytoplankton) as well as inelastic scattering in liquid water ("Ring effect"), leading to negative SCDs.
Fig. 2 shows the resulting mean Glyoxal SCD for 2003-2005 on a 1° resolution grid. So far, no further conversion to vertical column densities (VCDs) has been applied.
Applying a cloud filter does not change the observed patterns, but leads to increased noise and is therefore omitted yet.

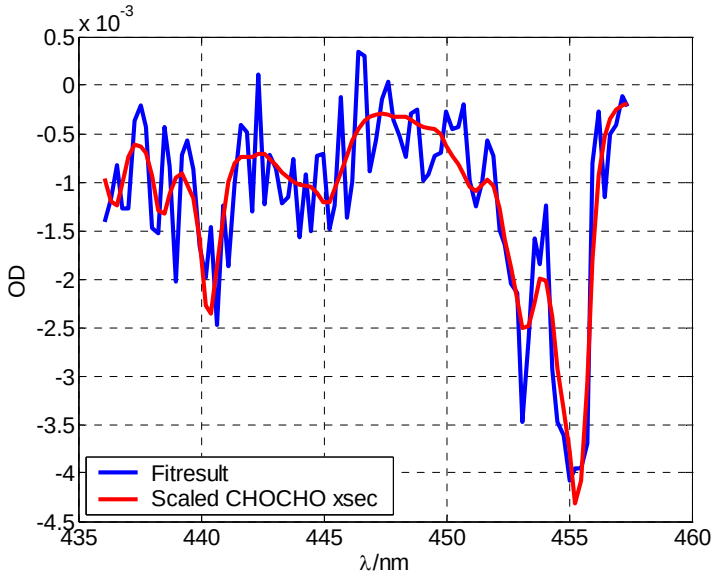

Fig. 1. Fit example: SCIAMACHY overpass over southeast Australia on 18 January 2003. The earthshine spectrum from the neighbouring southerly pixel was used as reference. The Glyoxal cross section was taken from [8].

3 RESULTS

In the mean Glyoxal SCD composite (Fig. 2), several anthropogenic sources show up (3.1). Furthermore, Glyoxal due to biomass burning can be identified (3.2). Also biogenic emissions can be observed, possibly even over some oceanic regions (3.3).

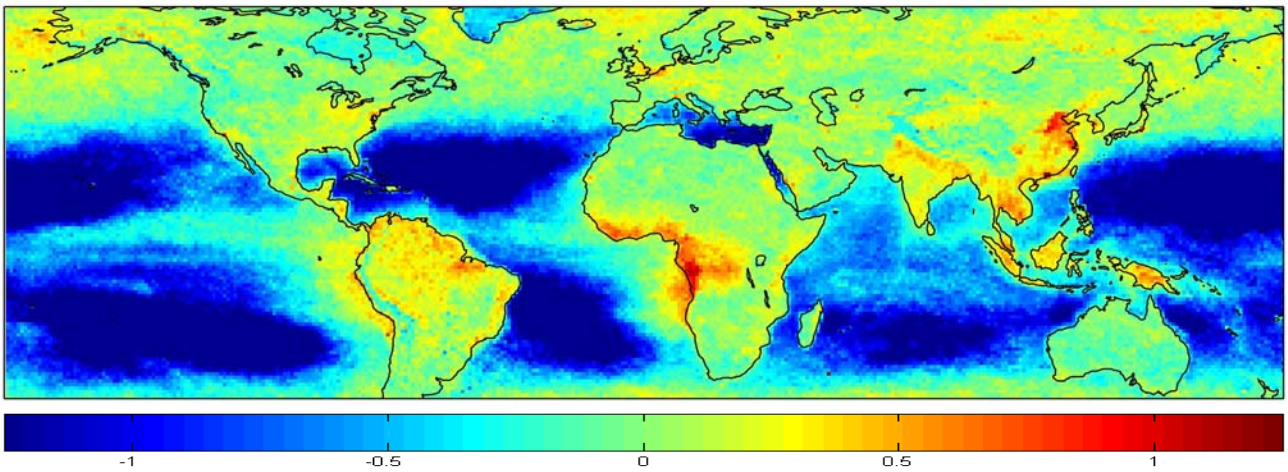

Fig. 2. Mean Glyoxal SCD 2003-2005. (10^{15} molec/cm^2)

3.1 Anthropogenic sources

Fig. 3 shows zooms of Fig. 2 for some regions of high anthropogenic activity. For comparison, the respective distribution of tropospheric NO_2 [7] is also displayed.

Several Glyoxal hotspots show up over large cities and congested areas where also very high tropospheric NO_2 VCDs are observed, for instance Los Angeles, US eastcoast (a), Belgium/Netherlands and the Po Valley (b), Hong Kong, Tokyo, Seoul and the Eastern China (c), or Rijad, Dschidda and the metropolises at the Persian golf (d). Hence photochemical hot spots due to anthropogenic activities can clearly be identified in the Glyoxal composite.

It has to be noted that this coincidence cannot be explained by a spectral interference what can easily be seen for fits for polar regions in summer, showing very high stratospheric NO_2 SCDs but no Glyoxal.

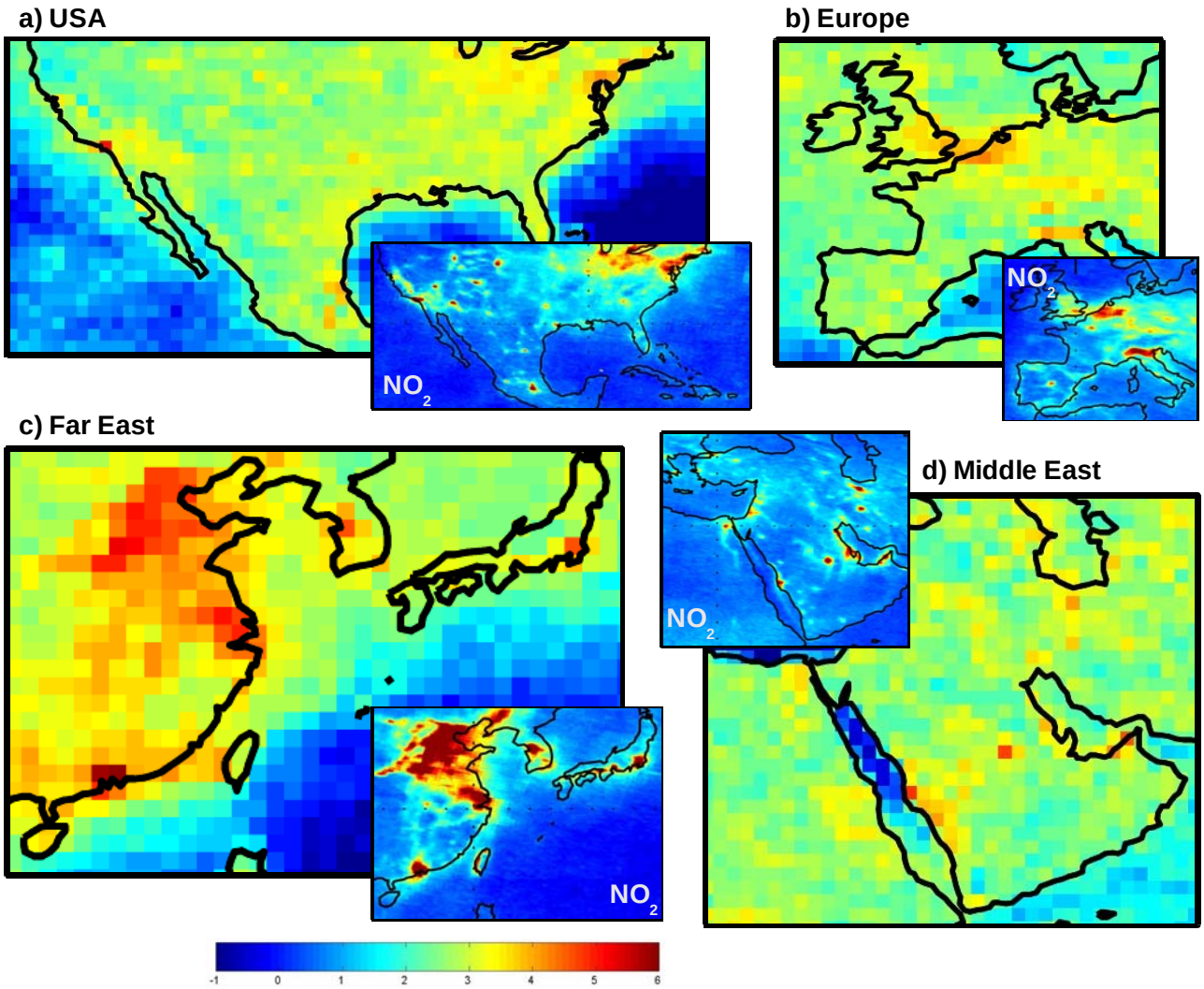

Fig. 3. Zoom of Fig 2 for (a) the USA, (b) Europe, (c) Far East and (d) the Middle East, showing enhanced Glyoxal SCDs due to anthropogenic emissions. For (d), the Glyoxal mean for 2003 is displayed. Colorbar as in Fig 2. The small images show the respective mean tropospheric VCD of NO_2 [8]. The colorbar for NO_2 is given in (c) (10^{15} molec/cm^2).

3.2 Biomass burning

Fig. 4 displays mean monthly Glyoxal SCDs compared to the respective ATSR fire counts over South America, indicating enhanced Glyoxal over biomass burning regions.

A direct coincidence of ATSR fire counts, enhanced tropospheric NO_2 and Glyoxal SCDs is shown in Fig. 5 for the disastrous fire storm on 18 Jan 2003 in southeast Australia.

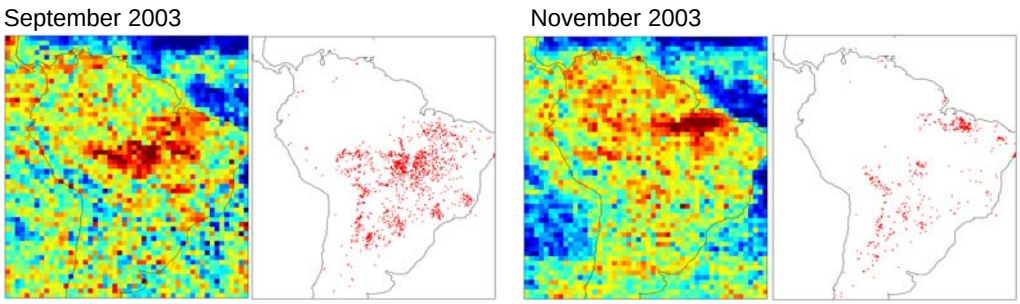

Fig. 4. Monthly mean Glyoxal SCD (left) and accumulated ATSR fire counts (right) for September and November 2003 over South America. Colorbar as in Fig. 2.

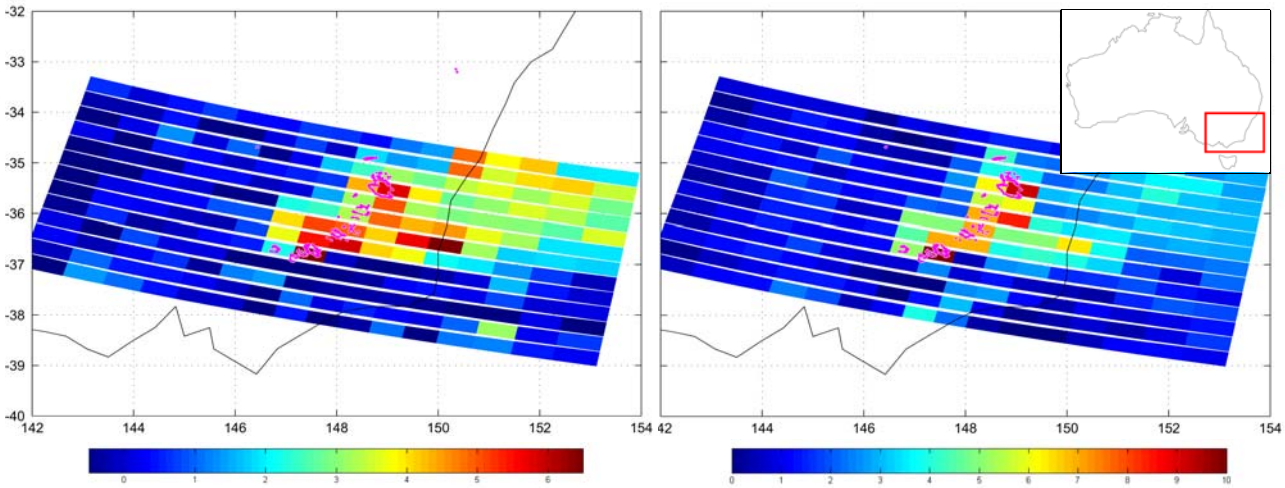

Fig. 5. (Left) Enhanced Glyoxal SCD (note the colorscale!) during biomass burning (magenta dots mark ATSR fire counts, total ~1000) in southeast Australia on 18 Jan 2003. (Right) Corresponding tropospheric NO_2 SCD.

3.3 Biogenic emissions – also over ocean?

In contrast to the examples shown above, fire counts and Glyoxal SCD do not correlate at all for August 2003 over Africa (Fig. 6). The very high Glyoxal SCDs at the western coast of Africa, and the generally high values over tropical rain forest, are probably due to biogenic emissions.

High Glyoxal SCDs are also observed over parts of the tropical ocean, e.g. west from Africa and South America (Figs. 2, 6). Due to the very short lifetime of Glyoxal this may indicate direct VOC emissions over these bioactive oceanic regions.

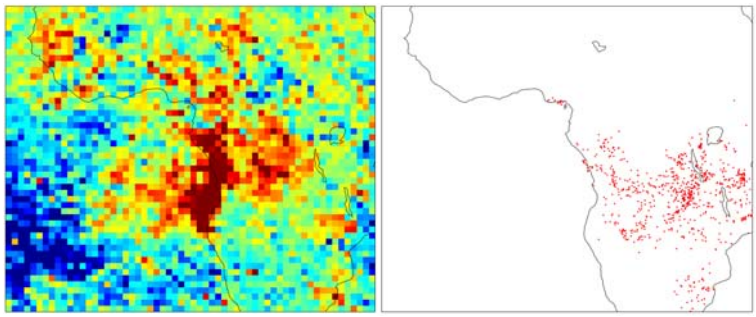

Fig. 6. Monthly mean Glyoxal SCD (left) and accumulated ATSR fire counts (right) for August 2003 over central Africa.

4 DISCUSSION AND OUTLOOK

Though causing comparably small optical depths, Glyoxal can be unambiguously detected from space. Photochemical hot spots due to anthropogenic emissions clearly show up in the 3 year composite map. Also enhanced Glyoxal levels due to biomass burning can clearly be identified. The high SCDs over tropical rain forest regions indicate biogenic emissions, and even emissions from ocean might have been detected. Ongoing SCIAMACHY measurements allow retrieving further information, e.g. on seasonal cycles, providing additional insight on Glyoxal sources.

To come to more quantitative statements, several issues have to be addressed. Further efforts have to be taken on the improvement of the fit, in particular over oceans, where the fit residuals have large structures and the fit results in negative Glyoxal SCDs. The impact of the solar reference on the fit quality and an overall offset has to be investigated further. For the conversion of SCDs in VCDs, additional information is needed, in particular on the Glyoxal profile. This depends on the fraction of Glyoxal being directly emitted or formed close to the ground and what fraction is produced from oxidation of (possibly long living) VOCs, what can also occur at higher altitudes.

The global time series of Glyoxal from satellite measurements allows comparisons with CTMs to check and improve our current understanding on VOC emissions and chemistry.

5 ACKNOWLEDGEMENTS

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6 REFERENCES

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