IDENTIFICATION OF TROPOSPHERIC TRACE GAS SOURCES: SYNERGISTIC USE OF HCHO AND OTHER SATELLITE OBSERVATIONS

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ABSTRACT

Satellite observations provide unique opportunities for the identifications of trace gas sources on a global scale. In our case, the satellite Formaldehyde (HCHO) observations provide information concerning the localization of biomass burning (intense source of HCHO over the Amazon basin region and in central Africa) and biogenic isoprene emissions. The HCHO data can be compared with NO_2, glyoxal and CO results to identify more precisely the tropospheric sources (biomass burning events, biogenic emissions, human activities).
We present case studies for combined satellite observations, derived from GOME measurements aboard the ERS-2 satellite and SCIAMACHY aboard ENVISAT as well as other satellite instrument results like fire counts.

1. INTRODUCTION

The identification of the source of tropospheric trace gas is a big challenge. We already know from the chemistry of each single trace gas the possible and expected sources. The problem comes from the overlap of several of those sources like for example biomass burning and biogenic emissions as sources for HCHO. The comparison of several trace gases from satellite observations allows to constrain more precisely the tropospheric sources of the trace gases.

2. METHOD

2.1. Instruments

Spectral measurements from the satellite instruments GOME (aboard ERS-2) and SCIAMACHY (aboard ENVISAT) allow to determine column densities of several important atmospheric absorbers, amongst them Formaldehyde (HCHO), Nitrogen dioxide (NO_2), Carbon monoxide (CO) and Glyoxal ($C_2H_2O_2$). For this comparison study we have also used the fires counts data provided by the instruments ATSR (aboard ERS-2) and AATSR (aboard ENVISAT).

2.2. HCHO retrieval

Maps of tropospheric HCHO columns were derived from spectroscopic data from the GOME instrument using the method of Differential Optical Absorption Spectroscopy (DOAS). The key concept of the DOAS method is the simultaneous fit of several trace gas absorption spectra to the ratio of the earth-shine spectrum and a Fraunhofer reference (Platt et al., 1994). For the retrieval we have used a daily calculate mean earthshine over the Pacific Ocean as Fraunhofer reference (Figure 1) instead of the daily measured sun reference spectra by GOME. This method allows to retrieve also the GOME data without direct sun reference measurements.

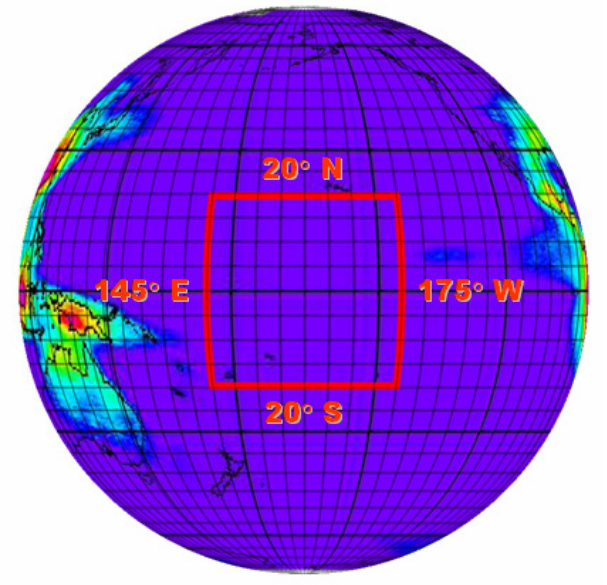

Figure 1. Localization of the daily calculated earthshine reference(145°E, 175°W, 20°N, 20°S)

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First, from the DOAS fitting procedure the so called slant column density (SCD, the trace gas concentration integrated along the light path) is calculated. In a second step, the radiative transport through the atmosphere is modeled. From such models the so called air mass factor (AMF) is determined, which is the ratio of the SCD and the vertically integrated trace gas concentration (VCD).
For HCHO we only calculated SCDs using an algorithm developed at the IUP Heidelberg, which is basically the same DOAS retrieval method as used for the HCHO ground measurements (Wagner et al., 2004). The stratospheric HCHO can be neglected. Thus, the retrieved slant columns directly represent the tropospheric HCHO. However, we applied a normalization to account for the discrepancies between the first and the last of the three forescan pixels from the GOME instrument. An Earth-shine ratio spectra between the first and the third pixel has been fitted with the HCHO retrieval to avoid this effect. A conversion to tropospheric VCDs will be performed in the near future. Nevertheless, since the most HCHO emissions are situated at low latitudes (small SZA), the spatial patterns are not strongly affected.

3. RESULTS

3.1. Biogenic Emissions

The 8 years HCHO results map (January 1996 – June 2003) gives an idea of the repartition of the HCHO anomalies (Figure 2). Three main HCHO sources are situated on the equator, on rain forests (Amazon basin, Africa and Indonesia). Weaker sources can also be observed over the south east of the US, over Mexico, Europe, India and in south west Asia.
The three main sources seen over the rain forest have been identified to be mostly due to biogenic emissions. This has also be demonstrated for the south east of the USA (Chance et al., 2000)

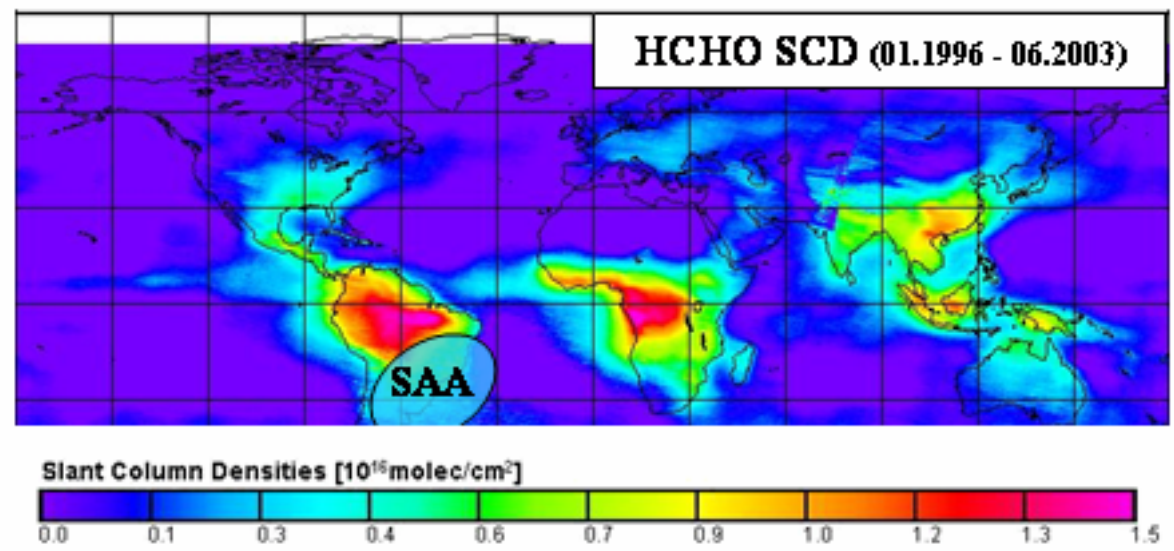

Figure 2. This global map of 8 years GOME HCHO SCD results shows the repartition of the enhanced HCHO column. The highest HCHO column densities are localized over the equatorial rain forest.

3.2. Biomass burning

Another important source for tropospheric HCHO is due to biomass burning. On figure 3, the comparison between fire counts from ATSR with GOME HCHO column densities over the same region shows a good agreement between both dataset. These examples have been taken from the particularly strong El-Niño years of 1997-1998. Dryer conditions around the Pacific Ocean have induced strong biomass burning over Indonesia in September 1997, Central America in May 1998 and also in the eastern part of Siberia during August 1998.
The biomass burning also occurs over region known to be strong HCHO biogenic sources. For example in September 1997, the HCHO emissions situated in the northern part of the Amazon basin region are not

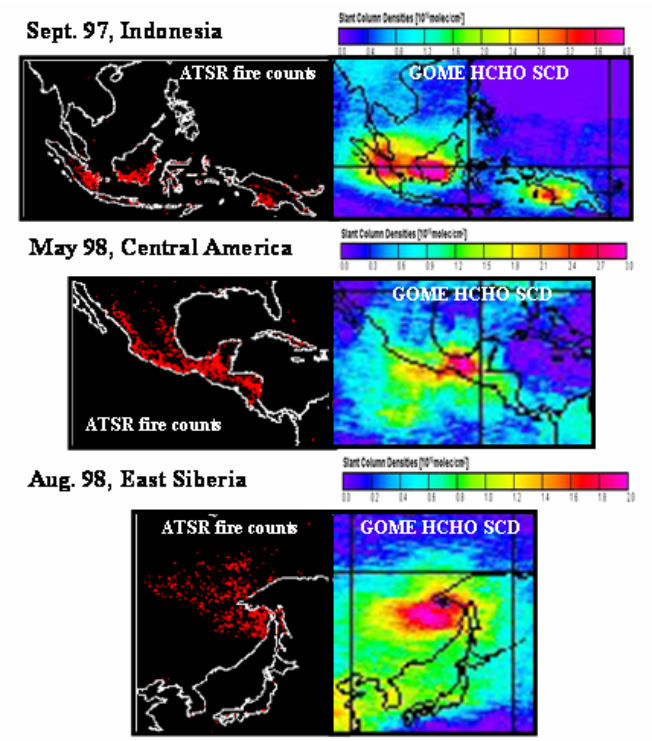

Figure 3. Fire counts due to Biomass burning correlated to the 1997-1998 El-Niño event. The fire counts are in good agreement with the HCHO SCD

correlated with forest fires (Figure 4). Only the southern part of the HCHO emissions correlates with the measured forest fires and also with the NO_2 concentrations. The northern part of the Amazon basin HCHO concentrations can be attributed to biogenic isoprene emissions. In this case study the NO_2 emissions are mostly due to the biomass burning and we know the part of HCHO due to isoprene emissions from the northern part. With these information we propose to discriminate the HCHO due to biogenic isoprene emissions from the HCHO due to biomass burning. Correlating with emission factors, we could apply this discrimination to places where no direct estimation from the HCHO part due to biogenic emissions can be made. Models could also help to identify the biogenic emissions and thus the contribution from each sources (biogenic and biomass burning) in the HCHO column density.

There seems also to be a dependence between the NO_2 emissions during biomass burning and the vegetation type: NO_2 correlate with HCHO over Africa (grassland fires) but not over Indonesia (forest fires). In South America, an augmentation of the NO_2 concentrations can be observed with the fire shift from the forest to grassland vegetation (Marbach et al., 2006).

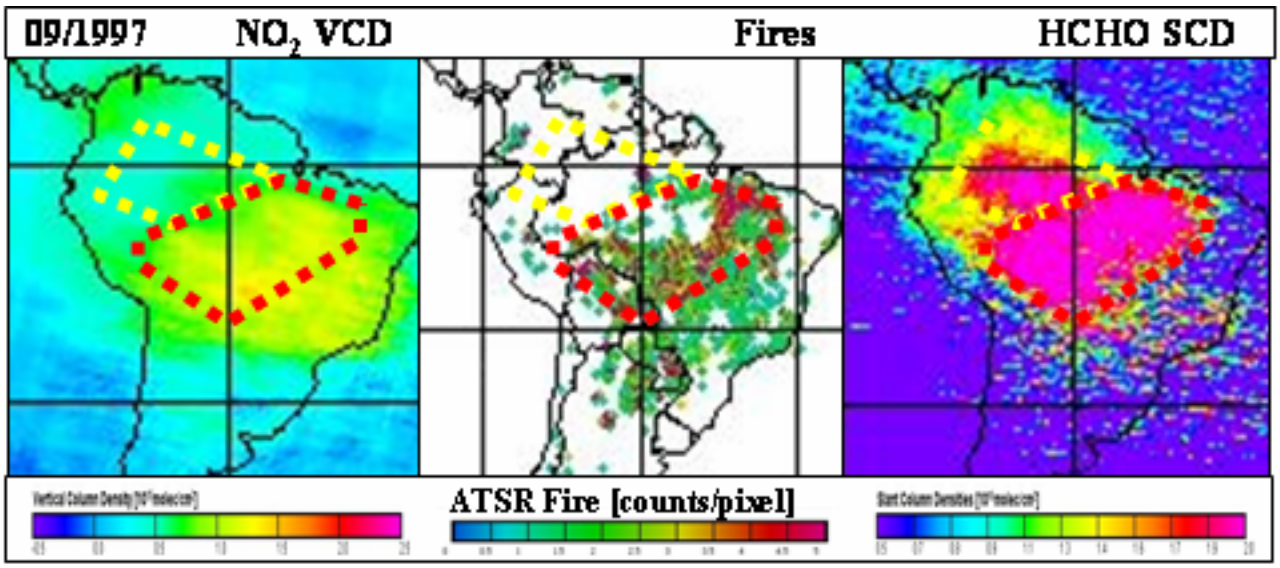

Figure4. In South America over the Amazon basin both sources (biogenic emissions as well as biomass burning) contributed to the HCHO columns densities measured by GOME. In the southern part of the basin (in red) both sources are added. In this region the fire counts are in good agreement with the NO_2 as well as with the HCHO tropospheric column densities .In the northern part(in yellow) only HCHO due to biogenic emissions can be observed (no fire counts and enhanced NO_2 columns in this region)

3.3. Synergistic use of HCHO, NO_2 and CO results

We propose to retrieve more precisely the tropospheric sources of the trace gases to use a combination of three trace gases (Figure 5). The advantage to use results from GOME and SCIAMACHY is that the measurements have been made around the same local time. The fires counts measured by AATSR are made during the night time of the satellite orbit, thus shifted to those from GOME and SIMACHY. CO could be in this way a proxy for the biomass burning. There is a good agreement over Africa between the CO VMR and the fires counts. The over enhanced CO VMR over China could be due also to anthropogenic activities and the NO_2 dataset could help to study this region. Finally the HCHO results will give information about the biogenic sources as well as the biomass burning sources (Part 3.2).
The region over China will be a good case study for the combination of CO, HCHO and NO_2 observations. The enhanced NO_2 tropospheric columns are situated in north east, the higher HCHO SCD in south east and the CO VMR seem to cover whole eastern China.

4. OUTLOOK

Another possible combination of trace gases is HCHO and Glyoxal. The mean global Glyoxal and Formaldehyde distributions indicate several photochemical hotspots due to anthropogenic emissions as well as biogenic and biomass burning VOC emissions. Also over some bioactive oceanic regions (Gulf of Congo or over the coast of Peru) enhanced Glyoxal levels correlate to enhanced Formaldehyde levels, possibly indicating VOC emissions over ocean. Both species are formed by the oxidation of several volatile organic compounds (VOCs) and hence serve as an important indicator for VOC chemistry. Both traces gases have approximatively the same lifetime of a few hours.

Statistical studies have to be made to validate the choice and the reality of the synergistic use of the trace gases to identify the tropospheric sources.

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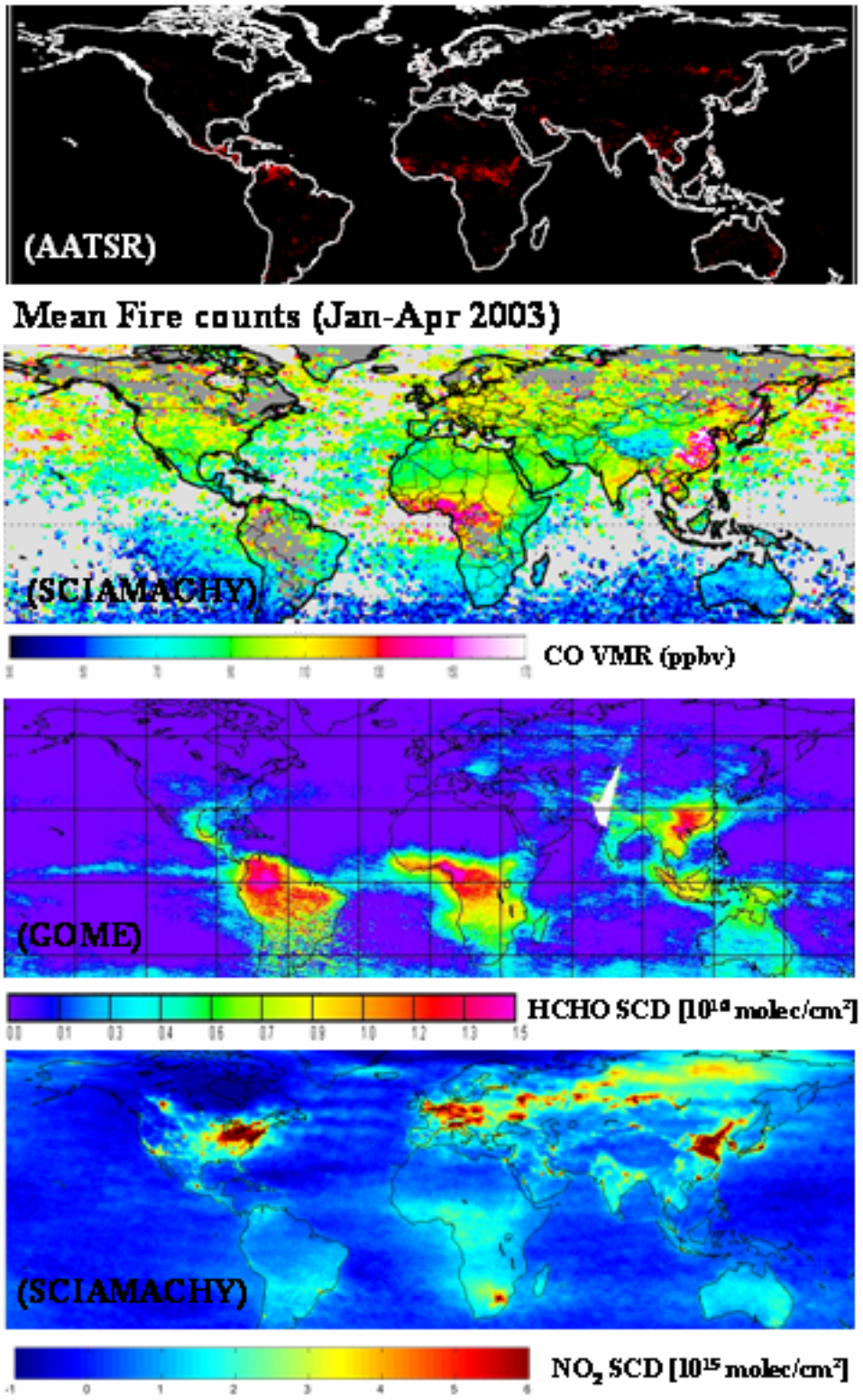

Figure 5. Mean maps (January-April 2003) of fires counts (AATSR), CO volume mixing ratio (SCIMACHY), HCHO SCD (GOME) and NO_2 SCD (SCIAMACHY)