GLOBAL OBSERVATIONS OF FORMALDEHYDE

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INTRODUCTION

Formaldehyde (HCHO) indicates and supports photochemical activity in the atmosphere. Large amounts are expected to found in industrial areas and during biomass burning. HCHO is a major intermediate in the degradation of methane (and many other hydrocarbons). In the absence of heterogenous losses, essentially every methane molecule is converted to HCHO. Therefore it is found throughout the troposphere. It is destroyed via photolysis and reaction with OH. In continental boundary layers, non-methane hydrocarbons (NMHCs) emitted by biogenic and anthropogenic sources dominate over methane also as a source of HCHO.

GOME is the first instrument, that allows observations of formaldehyde on a global scale giving the opportunity to improve our knowledge about emission fluxes of methane and NMHCs. In this study several months of all years since launch of ERS-2 in 1995 have been analysed for HCHO.

INSTRUMENT

The Global Ozone Monitoring Experiment (GOME) [2] observes the light scattered back from the atmosphere and reflected on the ground in near nadir viewing geometry. The GOME instrument is a grating pseudo double monochromator covering the spectral range of 240 to 790 nm with a spectral resolution of 0.2–0.4 nm. The ground pixel size is 320 km across-track and 40 km along track for the three forward scans and $960 \cdot 40 km^2$ for the back scan. Global coverage is achieved within three days at the equator and within one day at 67°. Once per day, an extraterrestrial solar spectrum is measured via a diffusor plate. The GOME instrument has been launched on the European ERS-2 satellite in April 1995 and is operational since July of that year. ERS-2 is in a sun-synchronous polar orbit with an equator crossing time of 10:30 UT.

The main objective of GOME is the determination of the global stratospheric ozone field. However, the broad spectral coverage and the relatively good spectral resolution also enables the retrieval of a number of other trace gases such as OClO, BrO and NO_2 with structured absorptions in the UV and visible wavelength range by the means of the DOAS algorithm. Under clear sky conditions, visible and near UV radiation reaches the surface, and provides GOME with a unique sensitivity towards tropospheric absorbers. In particular, GOME measurements have been used to study tropospheric SO_2 [4], BrO [12,8], and NO_2 [7]. In the case of formaldehyde some case studies have been done with small data sets [11, 5, 3].
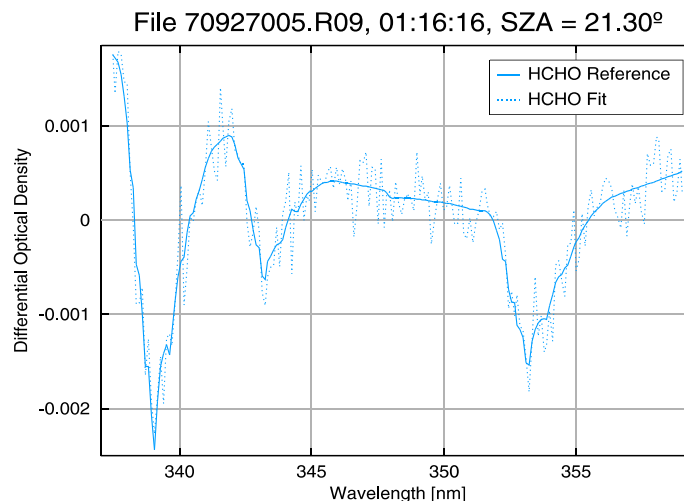

Fig. 1. One example for fitting of formaldehyde in GOME orbit 70927005. The thick blue line is the scaled laboratory reference, the dashed line is the residual after removing the contributions from scattering and other trace gas absorptions. The corresponding slant column density is $5.8 \cdot 10^{16}$ molec/cm^2. The linear fitting error is about 5 %.

ANALYSIS

Calibrated GOME lv1 earth-shine and solar measurements as provided by ESA have been analysed using the IUP Bremen Differential Optical Absorption (DOAS) algorithm to derive slant columns of HCHO. The wavelength window 337.5–359 nm is used for the fit as the interference by other species in particular ozone is relatively small. In addition to the HCHO cross-section [6], ozone (two temperatures), NO_2, BrO, O_4, and a synthetic Ring spectrum are included in the fit. Furthermore an undersampling correction and offset and slope correction functions have been applied to the fit. The solar spectra taken daily by GOME have been used as background spectra, minimising the effects of instrumental drift. The solar zenith angle range has been restricted to values below 75°.

Fig. 1 shows an example for the HCHO-Fit after removing all other contributions. The differential absorption corrresponds to a slant column density of $5.8 \cdot 10^{16}$ molec/cm^2. Fig. 2 shows the corresponding orbit crossing the biomass burning plume above Indonesia in September 1997.

Slant columns have been converted to vertical columns using airmass factors (AMF, Fig. 3) computed with the radiative transfer model SCIATRAN [9]. A simple standard tropospheric profile has been assumed for HCHO. This profile based on measurements performed during the TROPOZ II field campaign [1]. Chemical enhancement of AMFs has been neglected as well as the seasonal variation of atmospheric constitutions or variation of the albedo. For this study an albedo of 0.1 has been used for the whole hemisphere. This leads to an overestimation of the vertical column of HCHO under ice or snow conditions.

The cloud cover fraction derived by the operational ICFA algorithm is used, and only measurements below a treshold cloud fraction of 0.3 are included in the analysis (with the exception of September 2000). The derived vertical column data have been gridded to a resolution of 0.25°•0.5° (latitude, longitude).

In addition for selected scenarios other parameters have been used, providing information about biomass burning events. The ATSR product has been used to identify regions, where biomass burning takes place. The aerosol index derived from TOMS observations has been used to identify the extent of the biomass burning plume.
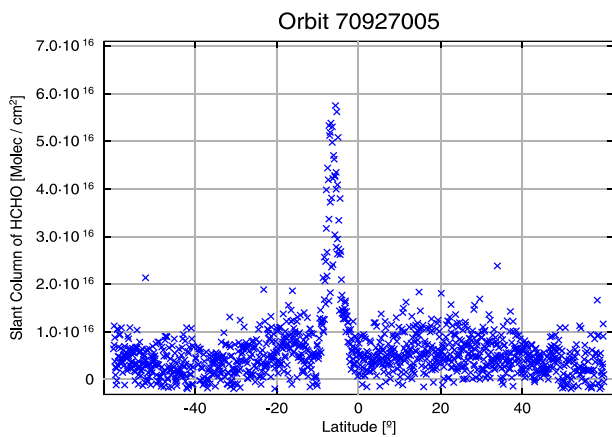

Fig. 2. The slant column densities derived for formaldehyde for an GOME orbit crossing the biomass burning plume in September 1997. See also Fig. 7 a.
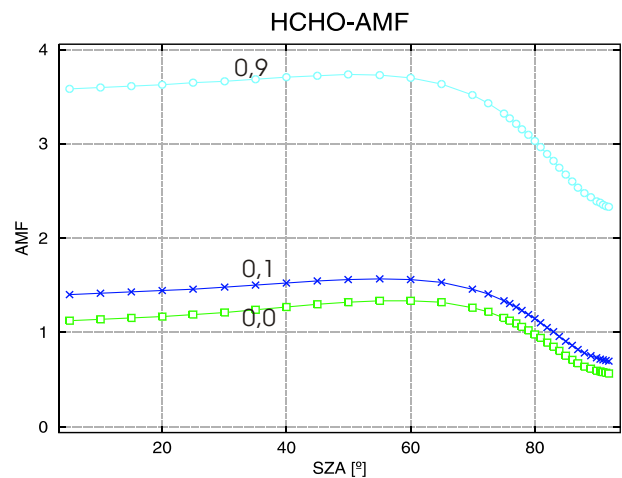
Fig. 3. The air mass factors as calculated by SCIATRAN [9] for different albedo for an tropospheric profile [1].

RESULTS AND DISCUSSION

The year-to-year variation as shown in Fig. 4 is quite small. The main pattern is very similar each year with large areas of enhanced HCHO amounts in South America and West Central Africa. These regions correlate quite well with fires as seen by ATSR (Fig. 6 b).

Slightly smaller values in 2000 (Fig. 4 f) are due to the fact, that the cloud selection was not possible for this month (no ICFA data available so far). High values of HCHO above the oceans (e.g. North Atlantic in 1995, Fig. 4 a) are probably due to interference with spectral effects on clear liquid water; in addition the albedo of water is usually higher than the albedo above the continents.

In September 1997 (Fig. 4 c) very high amounts of HCHO are visble above Indonesia. Related to the El Niño phenomena the monsoon rains expected during these months were absent, leading to very dry conditions and large biomass burning events [6,11]. The extent of the area with elevated HCHO levels is very similar to the TOMS observations of the biomass burning plume (Fig. 7 a).

Other biomass burning events in particular in other latitudes causes much smaller emissions of HCHO. This can be seen e.g. in September 1998 in East Siberia (Fig. 4 d and Fig 7 b). Other regions with smaller HCHO emissions are North Australia, North America and India, indicating that the emission of HCHO depends in a high degree on the material, which is burning (e.g. grasses in Australia or forests in North America) .

Figure 5 shows the seasonal variation of HCHO during 1999. Larger amounts of HCHO occur during summer season in each hemisphere. For March the correlation between biomass burning and elevated HCHO levels is much poorer as for September 1997 (Fig. 5 a and Fig. 6 b).

High values of HCHO above high latitudes (e.g. in Fig. 5 a and 5 d) are probably due to an albedo effect (snow and ice coverage, see AMF in Fig. 3), but sources in this regions can not ruled out in particular in the spring in each hemisphere [10].

Industrial sources of HCHO can be identified e.g. in Fig. 5 b (June 1999). Above the eastern part of North America high amounts of HCHO are probably due to emissions of the petrol industry. These sources will be analysed in more deatil in a forthcoming study.
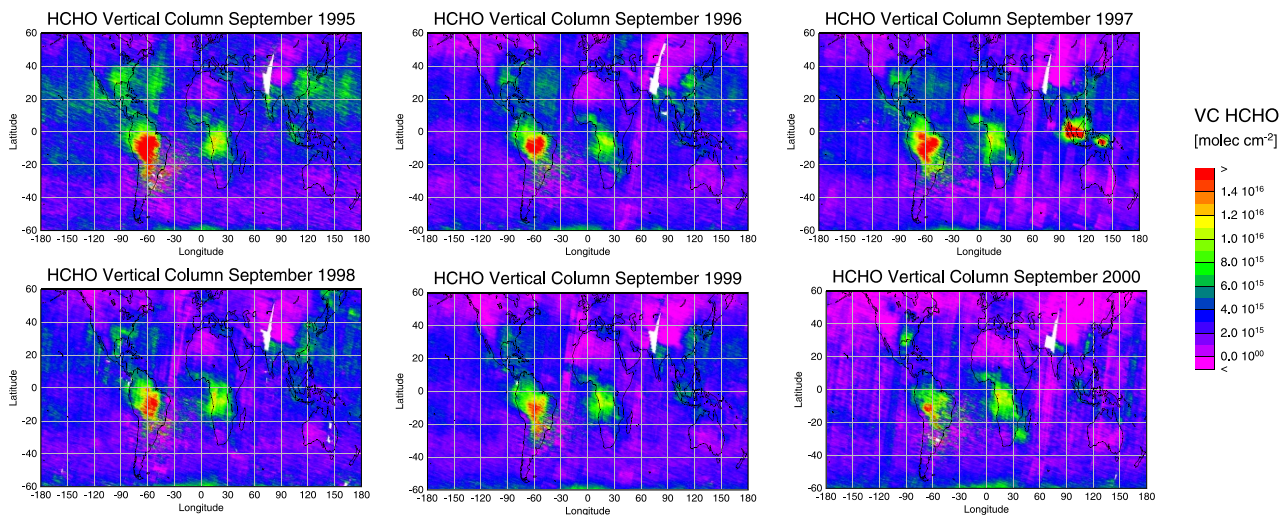

Fig. 4.a-f. Monthly means of HCHO vertical columns for each September from 1995 to 2000. The pattern is quite similar each year with the exception of September 1995 with high values above large areas of the oceans (see discussion).
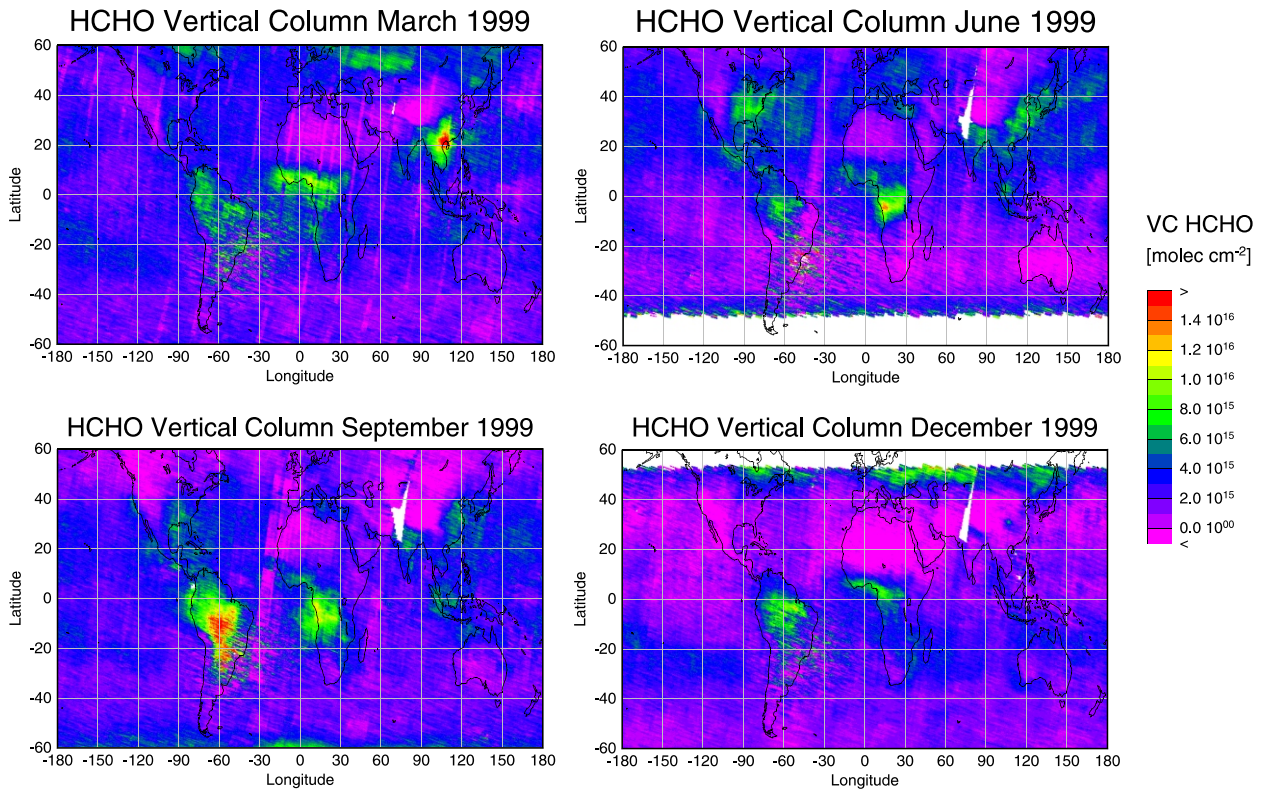
HCHO Vertical Column March 1999

HCHO Vertical Column June 1999

HCHO Vertical Column September 1999

HCHO Vertical Column December 1999

VC HCHO
[molec cm⁻²]

Fig. 5. a-d. Monthly means of HCHO vertical columns for different seasons in 1999.

Fires seen by ATSR in September 1997

Fires seen by ATSR in March 1999

Fires
[Counts]
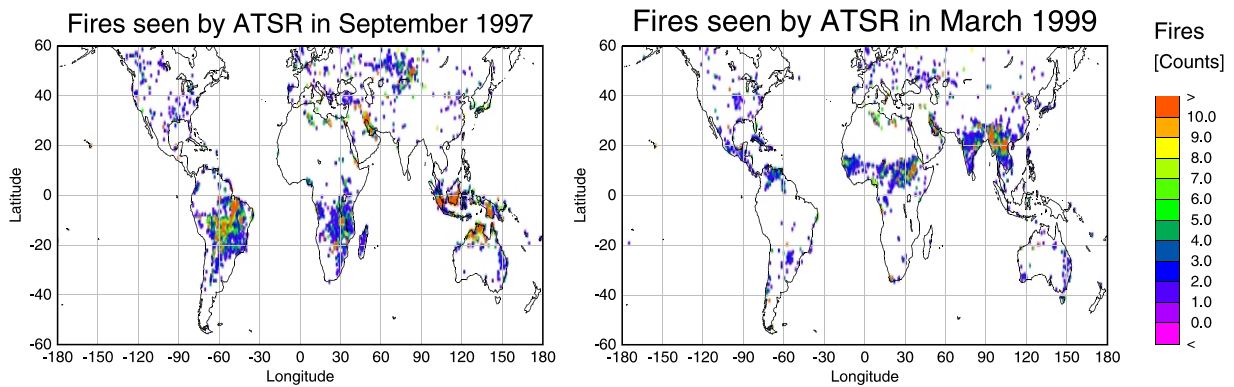
Fig. 6. a-b.The ATSR world fire atlas for September 1997 and March 1999 as provided by ESA.
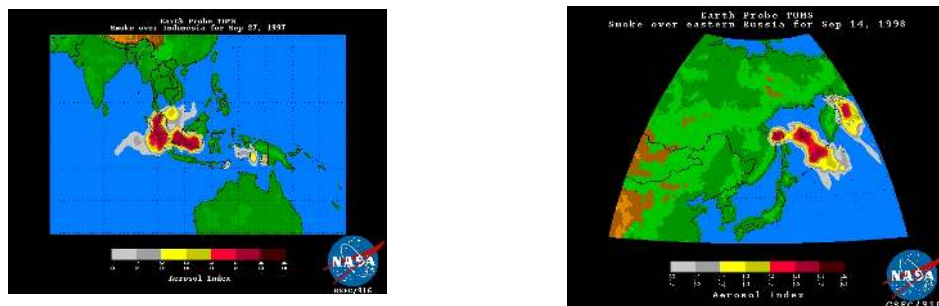

Fig. 7. a-b. The aerosol index as derived from TOMS observations.

OUTLOOK

In the near future our analysis will take into account the "right" albedo in the evaluation of vertical columns. A better correction of offset problems due to small changes in the solar irradiances used as background is another effect we will correct to get a more consistent picture of the global distribution of formaldehyde.

With the new generation of space borne UV/visible instruments such as SCIAMACHY on ENVISAT, better signal to noise ratio and smaller ground pixels will be possible. This should improve the accuracy of the HCHO columns.

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