Tropospheric CH_2O Observations from Satellites: Error Budget Analysis of 12 Years of Consistent Retrieval from GOME and SCIAMACHY Measurements.

A contribution to ACCENT-TROPOSAT-2, Task Group 1

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Introduction

Formaldehyde (CH_2O) is one of the most abundant hydrocarbons and plays a central role in tropospheric chemistry. Satellite measurements of CH_2O can be used to constrain VOCs emissions used in current state-of-the-art chemical transport models (see the contribution of Stavrakou et al., "10 years of pyrogenic NMVOC emissions deduced from CH_2O satellite data"). In order to perform reliable inverse modelling of emissions, consistent and properly characterized measurements covering several years are required. This work presents global tropospheric CH_2O columns retrieved from GOME and SCIAMACHY spectra. Efforts have been devoted to the assessment of the homogeneity of the data products derived from both platforms with the aim to derive a combined dataset of CH_2O vertical columns covering the 1996-2007 period. The retrieval has been performed using the same fitting interval and a consistent methodology for the evaluation of the air mass factors. A detailed comprehensive error analysis is presented. This includes errors on the slant columns retrieval and errors on the air mass factors which are mainly due to uncertainties in the a-priori profile, in the clouds properties and in the ground albedo.

Tropospheric CH_2O Vertical Columns

Figure 1 displays the GOME CH_2O vertical columns averaged over 7 years (from 1996 to 2002) and the SCIAMACHY CH_2O vertical columns averaged over the next 4 years (from 2003 to 2006).

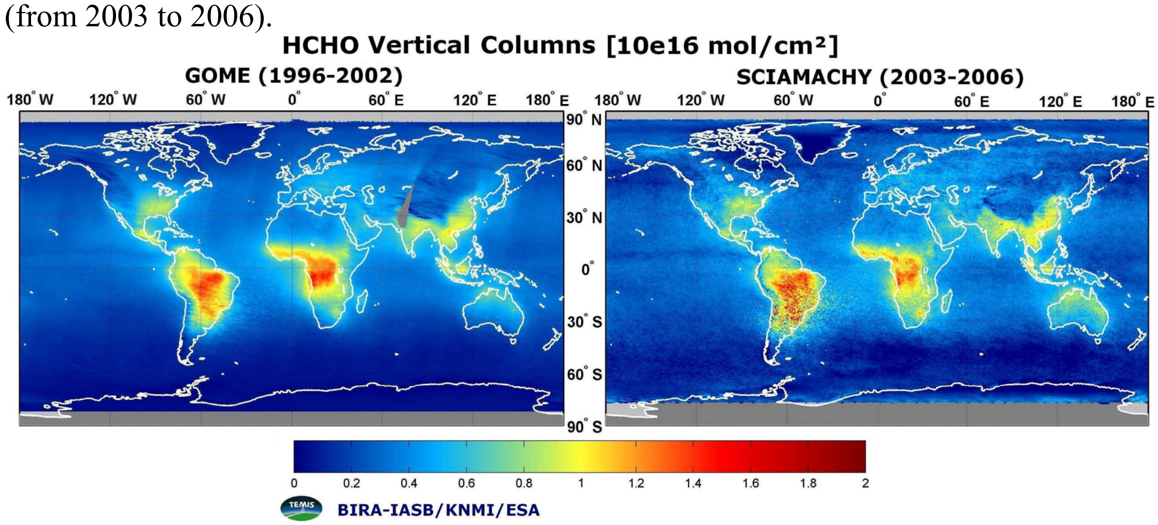

Figure 1: CH_2O vertical columns retrieved from GOME (1996-2002) and SCIAMACHY (2003-2006).

Vertical columns ***VC*** have been retrieved with the DOAS technique, using common settings for both instruments. ***VCs*** are defined as the ratio between the CH_2O slant column ***SC*** and the air mass factor ***AMF*** of the corresponding observation. Furthermore, a zonal correction is applied based on the background of CH_2O determined in a reference sector over the Pacific Ocean (SC_0 and VC_0).

$$VC = \frac{(SC - SC_0)}{AMF} + VC_0 = \frac{(\Delta SC)}{AMF} + VC_0$$

1. **Slant Column Fitting (*SC*)**

Slant columns of CH_2O are retrieved from GOME and SCIAMACHY spectra using the DOAS technique [Platt, 1994]. The wavelength interval used for the fit (328.5-346 nm) has been chosen carefully to improve the consistency between the two instruments and reduce the fitting artefacts in Tropical regions, particularly the abnormally low values in desert regions. The CH_2O absorption cross-sections applied in the fit are those of Cantrell [1990]. The fitting procedure also includes reference spectra for interfering species (O_3, NO_2, BrO, and O_4). The Ring effect [Grainger, 1962] is corrected according to Chance and Spurr [1997] using a solar irradiance measured by the satellite instrument as source spectrum. A linear intensity offset correction is applied as well as a polynomial closure term of order 5. Fraunhofer radiance spectra are selected on a daily basis in the equatorial Pacific Ocean, in a region where the formaldehyde column is assumed to be due only to methane oxidation and therefore small and stable in time. For more information see De Smedt et al. [2007].

2. **Air Mass Factor Evaluation (*AMF*)**

In the troposphere, total AMF depend on the scattering properties of the atmosphere, described by the weighting functions ***WF*** and on the vertical distribution of the molecule, described by the shape factor ***S*** [Palmer, 2001]:

$$AMF = \int_{atm} WF(z, angles, CF, CT, albedo, altitude).S(z, lat, long, month).dz$$

The weighting functions ***WF*** have been evaluated from radiative transfer calculations performed with a pseudo-spherical version of the DISORT code [Kylling, 1995]. The scattering properties of the atmosphere have been modelled for a number of representative viewing geometries, UV-albedos and ground altitudes. Albedos are taken from the climatology of Koelemeijer [2003]. The retrieval uses the cloud top height and cloud fraction data obtained using the FRESCO cloud product [Koelemeijer, 2002].
The shape factor ***S*** is the normalised profile of the absorbing molecule. Monthly CH_2O profiles are taken from the tropospheric chemistry transport model IMAGES [Müller, 2005] driven by analysed meteorological fields. IMAGES provides best-guess profiles of CH_2O, based on up-to-date emission inventories and a revised NMVOC chemistry mechanism optimized for the estimation of CH_2O chemical production.

3. **Background correction (SC_0 and VC_0)**

To reduce the impact of zonal artefacts in the slant columns, mainly due to ozone misfits in the CH_2O fitting window, an absolute normalisation is applied on a daily basis using the reference sector method [Khokhar, 2005] where the CH_2O background is taken from the tropospheric 3D-CTM IMAGES [Müller, 2005] in the Pacific Ocean.

Evaluation of the Error on the Tropospheric CH_2O Vertical Columns

As the determination of the ***SC***, ***AMF*** and VC_0 are independent, the total error on the tropospheric vertical column can be expressed as [Boersma, 2004]:

$$\sigma^2_{VC} = \left(\frac{\partial VC}{\partial SC}\right)^2 \sigma^2_{SC} + \left(\frac{\partial VC}{\partial AMF}\right)^2 \sigma^2_{AMF} + \left(\frac{\partial VC}{\partial VC_0}\right)^2 \sigma^2_{VC_0}$$

»» $$\sigma^2_{VC} = \frac{1}{AMF^2}\frac{\sigma^2_{SC\,rand}}{N} + \frac{1}{AMF^2}\sigma^2_{SCsyst} + \left(\frac{\Delta SC}{AMF^2}\right)^2 \sigma^2_{AMF} + \sigma^2_{VC_0}$$

Where σ_{SCrand} and σ_{SCsyst} are the random and systematic parts of the error on the slant columns, σ_{AMF} is the error on the air mass factor evaluation and σ_{VC0} is the error on the reference sector correction. The two last error sources are systematic. Contrary to the systematic error, the random error is reduced when the number of measurements increases. Therefore, σ_{SCrand} can be divided by the square root of the number of satellite pixels taken into the mean (N). These different error sources result in a total error generally comprised between 20 and 30% (Figure 2). They have been evaluated in detail for each observation conditions.

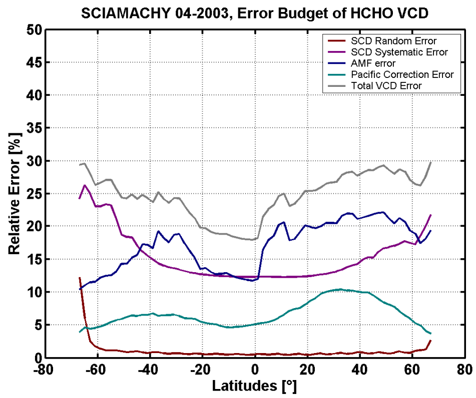

Figure 2: SCIAMACHY CH_2O VCD total error budget (averaged in April 2003).

1. Error on the CH_2O slant column (*SC*)

By definition, the random error on the slant columns is the standard deviation of the slant columns around the mean. It is well represented by the fitting error given in the DOAS fit (***SCDE***) if this ***SCDE*** is subtracted by its zonal dependency, particularly the systematic increase a high SZA due to ozone misfit (Figure 3). For GOME, σ_{SCrand} increases with years from 4e15 mol/cm² in 1996 to 6e15 mol/cm² in 2003 because of the degradation of the instrument. For SCIAMACHY, σ_{SCrand} reaches 1e16 mol/cm², because of the poorer signal to noise ratio due to the shorter integration time of this instrument. The random error on the slant columns is the most important source of error on the total vertical column but it can be reduced by averaging the data with a sufficient number of pixels.

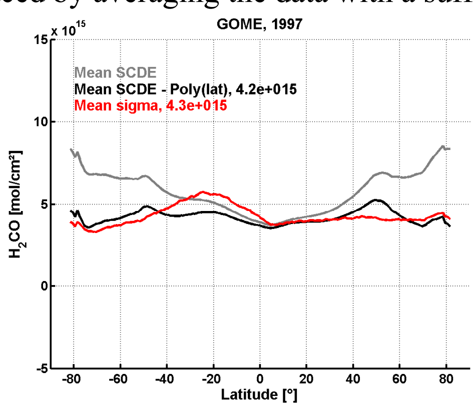

Figure 3: CH_2O SCD random error.

The systematic errors on the slant columns are due to errors in the slant columns fitting for example due cross-sections errors or interferences, nonlinearities, inaccurate calibration or missing corrections. The systematic errors due to cross-section errors and their correlations are calculated for each satellite pixel as a function of the concentration of each molecule included in the fit, following the optimal estimation theory [Theys, 2007]. An additional error of 12% accounts for other fitting parameter uncertainties. This value is based on sensitivity tests towards small changes in fitting window, calibration options or offset order. At high SZA, ozone and Ring absorption dominate the systematic slant column error. At lower SZA, CH_2O cross-section and fitting windows uncertainties have more importance.

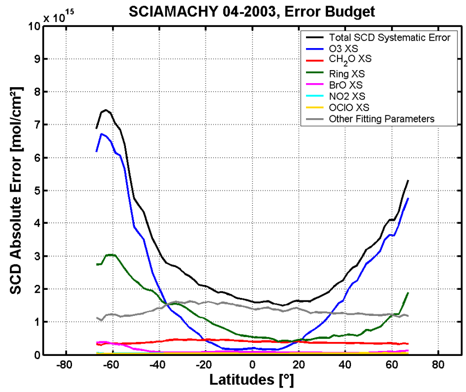

Figure 4: CH_2O SCD systematic error.

2. Error on the air mass factor (AMF)

The scattering properties of the atmosphere are modelled with weighting functions for representative values of the solar and viewing zenith angles and the ground albedo. The independent pixel approximation is used to correct for cloud effects, based on the cloud fraction and altitude of FRESCO. The vertical distribution of CH_2O is taken from the IMAGES model. The error on the total AMF depends on the uncertainties on all these parameters and on the sensitivity of the AMF to each of them:

$$\sigma^2{}_{AMF} = \left(\frac{\partial AMF}{\partial Alb} . \sigma_{Alb}\right)^2 + \left(\frac{\partial AMF}{\partial CF} \sigma_{CF}\right)^2 + \left(\frac{\partial AMF}{\partial CT} \sigma_{CT}\right)^2 + \left(\frac{\partial AMF}{\partial Sz} \sigma_{Sz}\right)^2$$

»» $$\sigma^2{}_{AMF} = K^2{}_{Alb} \sigma^2{}_{Alb} + K^2{}_{CF} \sigma^2{}_{CF} + K^2{}_{CT} \sigma^2{}_{CT} + K^2{}_{Sz} \sigma^2{}_{Sz}$$

The uncertainties on each parameter ($\sigma_{Alb}, \sigma_{CF}, \sigma_{CT}, \sigma_{Sz}$) have been taken from the literature and the sensitivities K_b have been evaluated for different representative conditions of observations and different profile shapes.

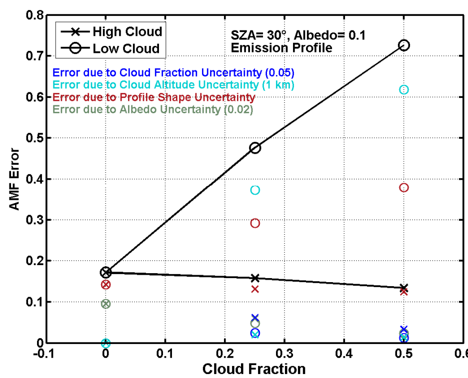

Figure 5: Contributions of each parameter to the AMF error in function of the cloud fraction for two cloud altitudes.

As shown in figure 5, the most important error sources are cloud altitude and profile shape under low cloud conditions (<2km). Other error sources like albedo and cloud fraction uncertainties have lower contribution to the total error on the AMF.

3. **Error on the background correction (VC_0)**

The uncertainty of the zonal correction has been evaluated by comparing the IMAGES CH_2O background over Pacific Ocean with the TM background in the same region. The differences are in the range of 0.5 to 2e15 mol/cm². Therefore, σ_{VCO} is small compared to other errors (see Figure 2).

Conclusion

A consistent dataset of CH_2O vertical columns has been created based on GOME and SCIAMACHY measurements. The quality and the consistency of this dataset is good enough to be used by the adjoin of the IMAGES model to provide top-down estimates of biomass burning and biogenic NMVOC emissions on the global scale. The satellite dataset is provided with a detailed error budget. For individual satellite measurement, the random error on the slant column is the largest source of uncertainty while monthly averages allow reducing this contribution to a negligible value. At high latitudes, the systematic error on the slant column dominates because of stronger ozone absorption. At lower and mid latitudes, the error on the air mass factor dominates with the main contribution from clouds and profile shape uncertainties. Pixels with cloud fractions above 40% and low cloud altitudes are characterized by very large errors (>50%) and should not be considered for quantitative analysis. The effect of aerosols on the error remains to be better quantified and could have an important impact on the retrieved CH_2O product in case of strong biomass burning events.

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