TREND DETECTION IN SATELLITE OBSERVATIONS OF FORMALDEHYDE TROPOSPHERIC COLUMNS USING GOME, SCIAMACHY AND GOME-2 SPECTROMETERS

Isabelle De Smedt, Michel Van Roozendael, Trissevgeni Stavrakou and Jean-François Muller
Belgian Institute for Space Aeronomy (IASB-BIRA), Belgium, isabelle.desmedt@aeronomie.be

Abstract

We present an update of the trend study published in (De Smedt et al., 2010). Global observations of formaldehyde columns consistently retrieved from the GOME, SCIAMACHY and GOME-2 measurements between 1997 and 2010 (***Fig. 1***) are linearly fitted using a trend model that also includes a seasonal component. The error and statistical significance of the inferred trends are estimated, taking into account the errors on the satellite observations (***Fig. 2***). Most significant changes are observed in Asia, where large positive trends are observed over Northeastern China (4% yr^{-1}) and India (1.8% yr^{-1}). In the United States, negative trends are found around several cities of the Eastern part of the country (New-York, Detroit, Dallas, Miami), illustrating the effective policies of emission reduction. However, significant positive trends are found over the Western part, around Los-Angeles and Phoenix (up to 4% yr^{-1}) (***Fig. 3)***. Using the seasonality of the H_2CO observations, we discriminate between the sources of the NMVOC emission trends (***Fig. 4 and 5***). The annual increases of the H_2CO columns in China and India are related to an increase of both the winter time columns, dominated by the contribution of anthropogenic NMVOC emissions, and the summer time columns, dominated by the biogenic source. In South-Western US, the annual increase of the H_2CO columns is due to an increase in the fall and winter time columns, directly related to anthropogenic emissions. However, in Eastern US, the negative trend in the winter time H_2CO columns (anthropogenic source) is cancelled be a positive trend in the summer time columns (biogenic source), which might be related to climate change effects.

Introduction

Atmospheric formaldehyde (H_2CO) is an intermediate product common to the degradation of many volatile organic compounds (VOCs). While the global formaldehyde background is due to methane oxidation, emissions of non-methane volatile organic compounds (NMVOCs) from biogenic, biomass burning and anthropogenic continental sources result in localised enhancements of H_2CO concentrations. Those can be mapped by recent satellite nadir sensors, helping to improve our knowledge of NMVOC emissions, which play an important role in global tropospheric chemistry and air quality. Albeit NO_x and NMVOC concentrations are expected to decrease in many industrialized countries, the rapid industrial development and urbanization in parts of Asia during the last decades resulted in significant increase of anthropogenic emissions, primarily from coal-fired power plants and automobile exhausts. In both cases, long-term measurements providing information on tropospheric trace gases are indispensable to assess and quantify these tendencies. Such time series can be used to monitor pollution trends and the climate change impact on the emissions.

Satellite Observations

A combination of H_2CO columns retrieved from GOME/ERS-2 (1997-2002), SCIAMACHY/ENVISAT (2003-2010) and GOME-2/METOP-A (2007-2010) is used in this study.

A complete description of the retrieval, based on the DOAS method, is provided in (De Smedt et al., 2008 and 2009). The retrieval settings have been homogenized as far as possible to avoid any inconstancies between the datasets. Figure 1 shows the mean H_2CO vertical columns as retrieved from GOME, SCIAMACHY and GOME-2 between 1997 and 2010. The use of the GOME-2 columns since 2007 allows for a reduction of the noise in the observations. Due to the decreasing sensitivity of tropospheric retrievals with increasing solar zenith angle, only columns for SZA lower than 60° are accounted for. Observations with cloud fractions larger than 40% are also filtered out. No explicit correction is applied for aerosols but the cloud correction scheme accounts for a large part of the aerosol scattering effect. The total error on the monthly and regionally averaged columns is comprised between 20 and 40% (De Smedt et al., 2008 and 2009).

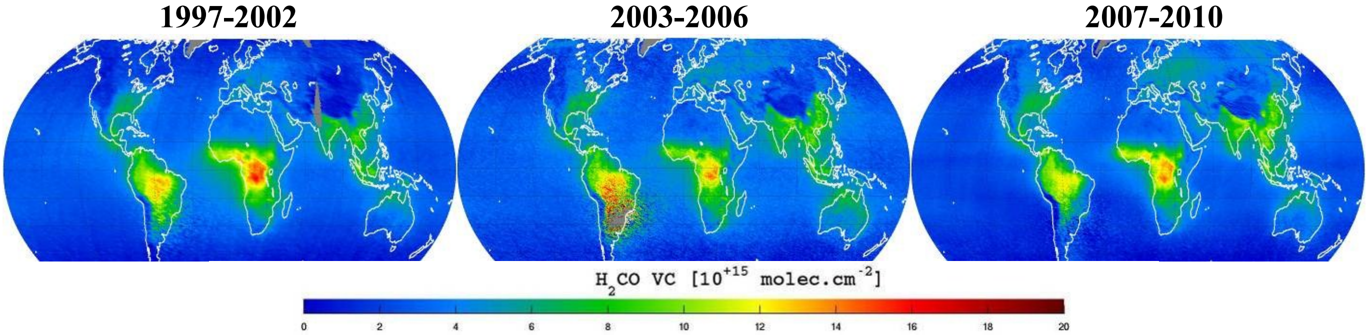

Figure 1: Mean H_2CO vertical columns between 1997 and 2010 as retrieved from GOME, SCIAMACHY and GOME-2 sensors.

Trend Analysis Method

To fit the time-series of monthly observations, a model with a linear trend and an intra-annual function, modelling the seasonal variations, has been used:

$$m(t) = A + Bt + \sum_{n=1}^{4} C_n \cos(n2\pi t) + \sum_{n=1}^{4} D_n \sin(n2\pi t) + U\delta$$

where ***m(t)*** is the observed monthly H_2CO ***VC*** for month ***t*** (expressed in fractional year), and ***A***, ***B***, C_n, D_n and ***U*** are the fitted parameters. ***A*** is the annual mean of the first year, ***B*** is the annual trend, expressed in molec.cm^2 yr^{-1}. C_n and D_n are the Fourier terms used to model the seasonal variations. ***U*** accounts for a possible bias between the satellite retrievals. A linear least-squares method is used to fit the model with the observations, taking their errors into account. In order to assess the statistical significance of the derived annual trends, the uncertainties on the fitted parameters (σ_B) are also estimated (De Smedt et al., 2010). The statistical significance level of the trend is better than 95% for $t_B = B/\sigma_B$ larger than 2.

The precision of the inferred trends is limited by the observation errors and by the length of the time series. To obtain significant results, the satellite monthly H_2CO columns need to be spatially averaged over relatively large areas, limiting the spatial resolution of the trend study. Figure 2 presents monthly averaged observations of H_2CO over India (left) and Beijing (right), and the fitted time series. The values of the annual trends ***B*** are respectively 1.2±0.3x10^{14} molec.cm^{-2} yr^{-1} and 1.4±0.4x10^{14} molec.cm^{-2} yr^{-1}, corresponding to annual growth rates of respectively 1.8±0.5% yr^{-1} in India and 4.0±1.2% yr^{-1} in the Beijing area.

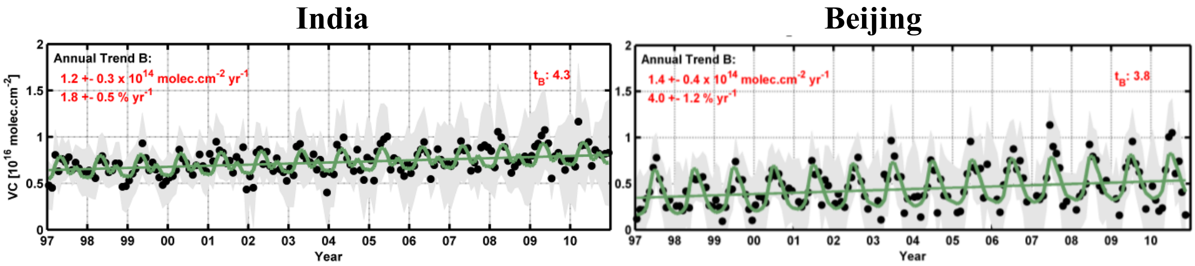

Figure 2 : Monthly averaged H_2CO VC (black dots) retrieved from GOME, SCIAMACHY and GOME-2 between 1997 and 2010, and fitted time series (green lines). Values of annual trends are indicated in red.

Annual and seasonal trends in Asia and the United States.

To draw a map of the inferred trends in Asia, monthly averaged time series of H_2CO observations have been calculated by country, and by province or state in China and India. Additionally, a trend study was performed using monthly averaged H_2CO data around largest urban areas of the United States. Figure 3 presents the results for Asia (left panel) and the US (right panel).

China and India are the countries presenting the most significant trends, of respectively 3.0±0.9% yr^{-1} and 1.8±0.4% yr^{-1}. Over China, the trend is larger in the Northeastern part, *i.e.* in the region with the highest anthropogenic emissions, and reaches 4% yr^{-1} in the Beijing area, whereas in Southeastern China the trend is lower (1.4% yr^{-1}). In Northern China, the sparsely populated province of Inner Mongolia shows a strong trend of 6.2±2.5% yr^{-1} between 1997 and 2010, whereas the Ningxia province, bordering Inner Mongolia, has the highest relative trend of 8±3.5% yr^{-1}, indicative of a very fast industrial development most likely related to the construction of new power plants [*Zhang et al.,* 2009]. India exhibits spatially homogeneous trends ranging between 1 and 2% yr^{-1}. In the Eastern United States, we find significant negative trends of about -2 % yr^{-1} over New York, Washington, Dallas, Philadelphia and Miami. More surprisingly, Los Angeles and San Francisco present positive annual growth rates of 2 and 3% yr^{-1}.

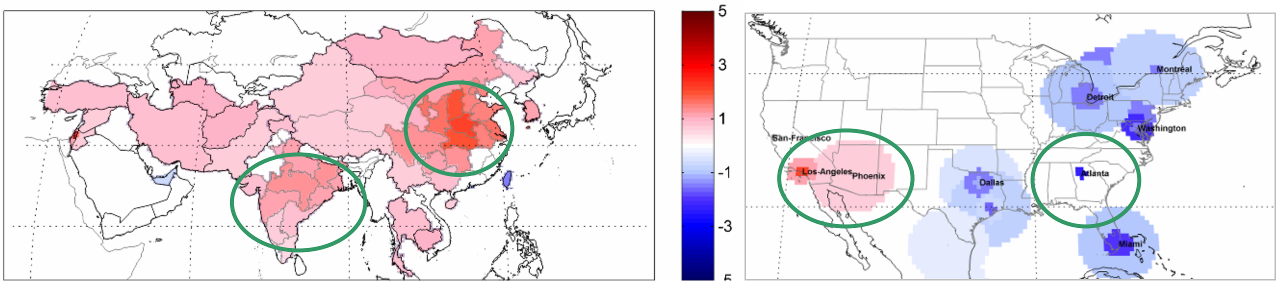

Figure 3: Maps of significant annual trends in H_2CO columns calculated around large American cities and in Asian countries between 1997 and 2010 (in 10^{14} molec.cm^{-2} yr^{-1}). Blank areas represent not statistically significant results. Green circled are the regions selected to study seasonal trends (see Figures 4 and 5).

Using the seasonality of the H_2CO observations, we attempt to discriminate between the sources of the NMVOC emission trends in the 4 regions circled in green in Figure 3. Figures 4 shows the seasonal trends calculated over India and Eastern China. In both regions, trends are strongest during winter and summer periods, reflecting respectively an increase of anthropogenic and biogenic emissions. The H_2CO trends are spatially consistent with changes in the REAS anthropogenic NMVOC emission inventory. However, simulations performed with the IMAGESv2 model suggest that the increase of the NMVOC emissions might be underestimated in REAS (De Smedt et al., 2010).

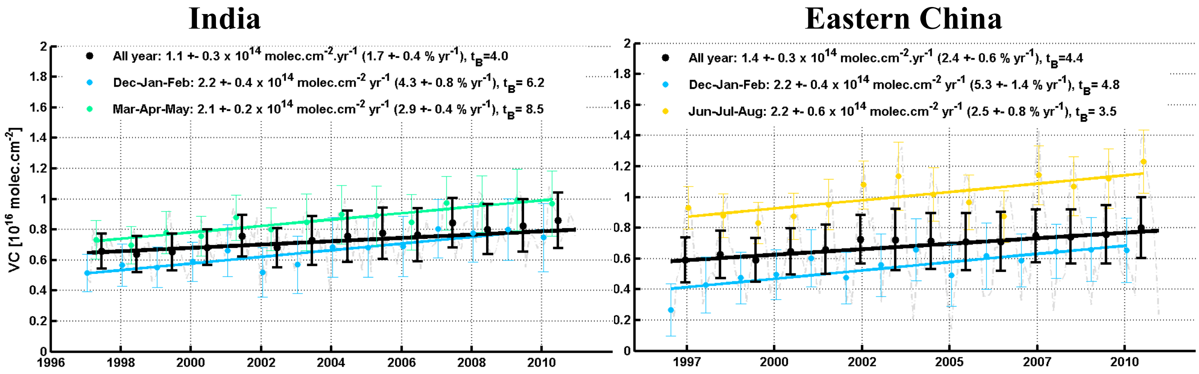

Figure 4: Seasonal H_2CO trends calculated over India and Eastern China

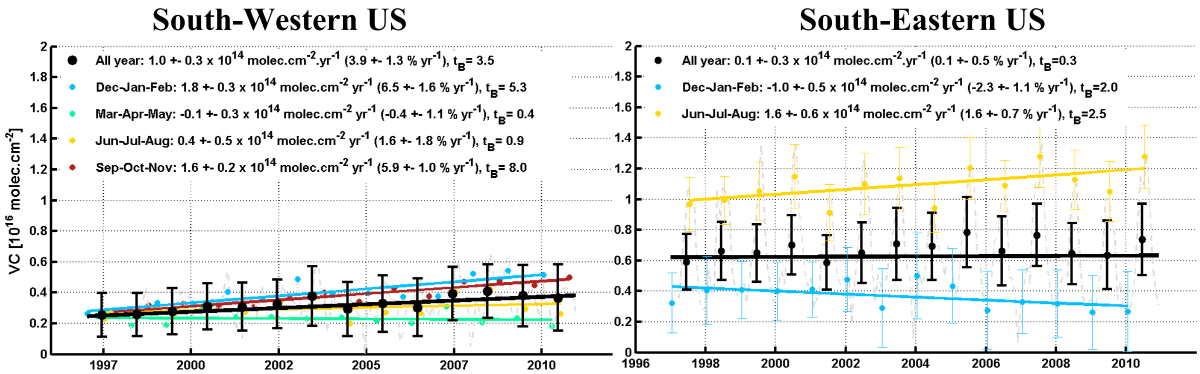

Figure 5: Seasonal H_2CO trends calculated over South-Western and South-Eastern US.

Figure 5 shows the seasonal trends calculated over South-Western US (left panel), where an annual increase is observed, and South-Eastern US (right panel), where the negative trends found over Eastern US cities is difficult to detect (see also the right panel of Figure 3). In SW US, positive trends are found in fall and winter time, when the contribution of anthropogenic sources is strongest. In SE US, the expected negative wintertime trend due to anthropogenic emission reduction policies is observed (-2.3 % yr^{-1}) but seems to be cancelled by a positive trend in summer H_2CO columns (+1.6 % yr^{-1}), caused by biogenic emissions over this densely forested region. The latter result, which could be related to climate change effects, needs to be further investigated and compared with modelled biogenic NMVOC emissions (MEGAN).

References

De Smedt, I., Müller, J.-F., Stavrakou, T., van Der A, R., Eskes, H. and Van Roozendael, M.: Twelve years of global observation of formaldehyde in the troposphere using GOME and SCIAMACHY sensors, Atmos. Chem. Phys., 8(16), 4947-4963, 2008.

De Smedt, I., T. Stavrakou, J.-F. Müller, N. Hao, *et al.* (2009), H_2CO columns retrieved from GOME-2: first scientific results and progress towards the development of an operational product, *Proceedings of the 2009 EUMETSAT Meteorological Satellite Conference*, Bath, U.K.

De Smedt, I., Stavrakou, T., Müller, J.-F., van Der A, R. J. and Van Roozendael, M.: Trend detection in satellite observations of formaldehyde tropospheric columns, Geophys. Res. Lett., 37(18), L18808, doi:10.1029/2010GL044245, 2010.