Constraining European biogenic isoprene emissions using satellite observations of formaldehyde

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Abstract – Formaldehyde (HCHO) is an intermediate product of the atmospheric oxidation of many volatile organic compounds (VOCs). While long lived VOCs such as methane provide background levels of HCHO, biogenic isoprene is believed to play a primary role in the spatial and temporal variability of HCHO, because of its high reactivity and high HCHO formation potential. Reactive anthropogenic VOCs may also significantly contribute to HCHO variability close to large sources. The new OMI product of HCHO column is a promising tool for VOC emissions estimate from space. We present the first comparison of OMI HCHO column with chemistry transport model simulations in summer. The model is used to estimate a quantitative relationship between isoprene emissions and formaldehyde column over Europe, then used to invert OMI HCHO column for isoprene emissions. Differences with a priori are evaluated against available independent VOC measurements at the ground.

Keywords: formaldehyde, biogenic emissions, air quality, chemistry transport model, VOC.

1. INTRODUCTION

Non-methane hydrocarbons, or volatile organic compounds (VOCs), contribute to the evolution of the oxidizing capacity and the optical properties of the atmosphere, through the formation of ozone and secondary particulate matter (Finlayson-Pitts and Pitts, 1997). Previous work showed that HCHO concentrations can be used to estimate emissions of VOCs (Palmer et al., 2003). The efficacy of this approach in determining the emission of a particular VOC depends on two factors: 1) the parent VOC having a significant HCHO yield *and* 2) the parent VOC having sufficiently short lifetimes such that there exists a local relationship between the emission of the VOC and the observed HCHO column. Palmer et al. (2003, 2006) illustrated this approach for isoprene by using column observations of HCHO from the Global Ozone Monitoring Experiment (GOME), in combination with the GEOS-Chem model, over North America during summertime when isoprene explained most the observed variability of the column. Shim et al. (2005) extended the isoprene emission inversion to the global scale at a lower resolution, while Barkley et al. (2008) applied the method to the Amazonian forest. The only study specifically focusing on Europe is by Dufour et al. (2009), who examined HCHO column measured by SCIAMACHY and retrieved correction factors to isoprene emissions at several European subdomains.

Unlike the global scale, in Europe AVOC emissions exceed BVOC emissions on yearly basis (Simpson et al., 1999), but the latter may still play an important role in summer, when BVOC emissions are at their maximum. In a recent multi-year assessment, Steinbrecher et al. (2009) reported that 40% of European BVOC emissions are concentrated in July, almost equally shared among isoprene, terpenes and OVOCs. June and August contribute with an additional 25% of the emissions each. During the vegetation growing season (April to September) BVOCs contribute about 2.5 ppbv to average surface ozone maximum over continental Europe, with peaks of 15 ppbv and 5 ppbv respectively over Portugal and the Mediterranean basin (Curci et al., 2009). Moreover, BVOC emissions were reported to contribute 30-75% to ozone production during specific severe events (Duane et al., 2002; Solmon et al., 2004). In contrast, BVOCs lead to a net ozone loss through the year in Northern European boundary layer (Curci et al., 2009). The uncertainty related to modelling of European BVOC emissions at small scale is estimated in a factor of 2-3 for isoprene and a factor of at least 5 for monoterpenes (Simpson et al., 1999; Steinbrecher et al., 2008). This yield an uncertainty in the estimate of the impact of BVOC on ozone levels of about 50% (Curci et al., 2009).

The final aim of this work is to exploit satellite observations of formaldehyde column from OMI to constrain isoprene emissions over Europe. Here, we examine the monthly mean HCHO column observed by OMI, and try to interpret its spatial distribution in relation to the emission of its precursors

2. ANALYSIS HCHO COLUMN OVER EUROPE IN 2005

2.1 Observations from OMI

The Ozone Monitoring Instrument (OMI) (Levelt et al., 2006) is one of the detectors mounted on the EOS-Aura spacecraft (http://aura.gsfc.nasa.gov/), launched in July 2004 on a near-polar sun-synchronous orbit. In the daytime ascending direction, Aura crosses the equator around 13:45 local time (LT), letting OMI observing Europe between 10:00 and 14:00 UTC. The instrument detect Earth's backscattered radiation with two CCDs in the UV/Vis spectral range (270-500 nm), scanning the atmosphere in the nadir direction with 60 across-track pixels along a swath of 2600 km that permits a near-global coverage in one day. The maximum pixel resolution is 13 × 24 km^2 at nadir and degrades toward swath edges.

Here we use the OMI 1-Orbit level 2 swath HCHO product (version 003, algorithm version 2.0) publicly released in May 2008 through NASA's DAAC (http://disc.gsfc.nasa.gov/). The HCHO column is retrieved by direct fitting of radiances and irradiances in the 327.5-356.5 nm UV spectral window (OMI-

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ATBD, 2002). The uncertainty on a single HCHO slant column observation ranges 40-100%, with lower end over hot-spots (Chance and Kurosu, 2008).

In Figure 1 we show the seasonal cycle of HCHO slant column observed by OMI in 2005. A seasonal cycle with a winter minimum, a summer maximum, and qualitatively in phase with the growing season (April to September) is clearly visible from OMI data. The column is below 6-8 × 10^{15} molecules cm^{-2} until May, when two slightly enhanced features above the Po Valley (Northern Italy) and Benelux appear. Over the continent, HCHO column is mostly > 8 × 10^{15} molecules cm^{-2} from June to September and mostly > 10 × 10^{15} molecules cm^{-2} in July. From October the column returns to low winter values.

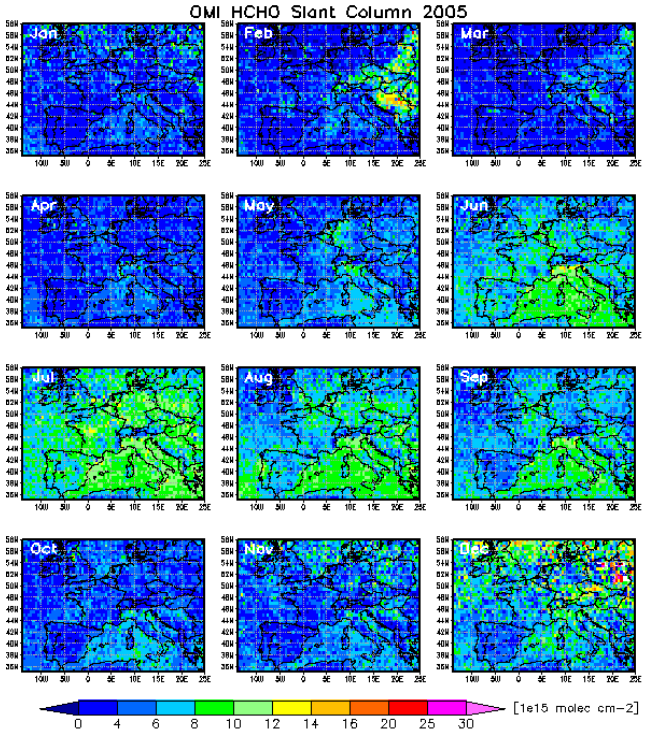

Figure 1. Monthly averages of HCHO slant column for year 2005 retrieved from OMI (on-board the EOS-Aura satellite), mapped onto a 0.5° × 0.5° regular grid over Europe.

2.2 Factors affecting HCHO variability

In Figure 2 we show the monthly mean HCHO column observed by OMI over Europe in summer 2005. Isoprene is likely to play a key role on HCHO column variability in summer over Europe. Isoprene emissions are related to temperature in an exponential fashion. Also photochemical activity is strictly related to temperature. We thus include in our analysis maps of temperature anomalies in Figure 2. The source of the monthly gridded fields of temperature anomalies is the Climatic Research Unit[1] in UK. Temperature anomalies are calculated with respect to the 1961-1990 period. Although it might be expected that temperatures in 21st century display always a positive anomaly with respect to that period because of global warming, the picture coming out is still useful to evaluate inter-annual and inter-monthly variability of temperature. In Figure 2 we can see that June 2005 was much warmer than usual with respect to July and August 2005, especially in West-Southern Europe.

Also fires are known to be a source of formaldehyde and other VOCs (Andreae and Merlet, 2001). We calculated total number of fire counts on a 0.5°x0.5° grid from the ATSR World Fire Atlas[2]. In Figure 2 we display results for June-August 2005. A large number of fires, potentially affecting HCHO column variability, is observed only over North Portugal during this period. A minor effect is also expected in the South of Italy.

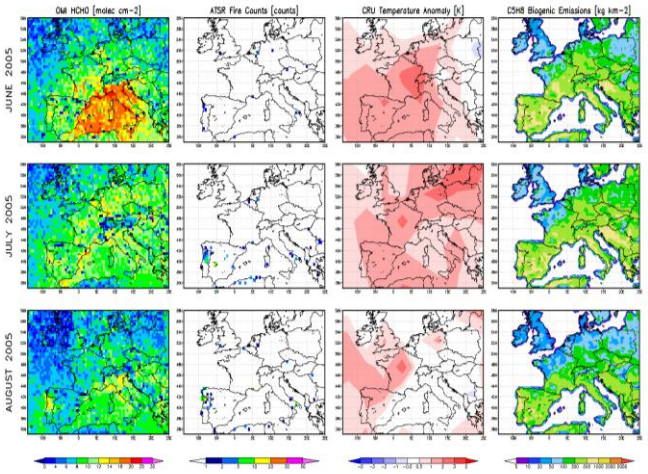

Figure 2. Summary of selected factors driving formaldehyde column over Europe. First column from the left: monthly average HCHO column observed by OMI in summer 2005 (June to August from first to third row) remapped on a 0.5°x0.5° grid. Second column: number of fires per month detected by ATSR on same grid as first column. Third column: monthly temperature anomaly calculated by CRU on a 5°x5° grid. Fourth column: total isoprene emissions calculated with MEGAN on same grid as first column.

3. CONCLUSIONS

This preliminary analysis of the HCHO column over Europe confirms the potential of the OMI product as a constraint for isoprene emissions. Indeed, the seasonal cycle appears to be driven by temperature and related emissions from vegetation. In the poster for the conference, final results of the comparison with model results and the inversion will be presented.

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[1] CRU, http://www.cru.uea.ac.uk/cru/data/temperature/. In Figure 2 the variance adjusted combined land-ocean temperature anomalies are displayed (product HadCRUT3v).

[2] http://dup.esrin.esa.int/ionia/wfa/index.asp

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