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Seasonally resolved global distributions of glyoxal and formaldehyde observed from the Ozone Monitoring Instrument on EOS Aura

Thomas P. Kurosu, Kelly Chance, Xiong Liu [1]
Rainer Volkamer [2]
Tzung-May Fu, Dylan Millet, Daniel J. Jacob [3]
Pieternel Levelt [4]

[1] Harvard-Smithsonian Center for Astrophysics
60 Garden Street, Cambridge, MA 02138, USA
{tkurosu, kchance, xliu}@cfa.harvard.edu

[2] University of California at San Diego
500 Gilman Drive, La Jolla, CA 92093, USA
rainer@chem.ucsd.edu

[3] Harvard University
29 Oxford Street, Cambridge, MA 02138, USA
{fu, millet, djacob}@fas.harvard.edu

[4] Royal Netherlands Meteorological Institute
Wilhelminalaan 10, PO Box 201, 3730 AE De Bilt, The Netherlands
pieternel.levelt@knmi.nl

Abstract. We present the first-ever analysis of seasonally resolved global distributions of glyoxal (CHOCHO) and formaldehyde (HCHO). These are simultaneously observed from the Ozone Monitoring Instrument OMI) on EOS Aura. Both gases are oxygenated volatile organic compounds (OVOCs) that are intimately linked to air quality monitoring. A 12-month time series of CHOCHO and HCHO total column monthly means during 2005-2006 is analyzed, focusing on regions of biomass burning (the Amazon, Africa, South-East Asia, etc.) and anthropogenic sources, in particular megacities including Los Angeles, Mexico City, the Pearl River Delta (PRD), and Beijing. OMI observations are compared with selected ground-based measurements from the MILAGRO and PRD campaigns, as well as with global simulations from the GEOS-Chem chemical transport model.

Keywords: remote sensing, air quality, volatile organic compounds (sensoriamento remoto, qualidade ar, complexo volatil organico).

Extended Abstract

1. Scope and Status of the Study

This study combines satellite measurements of CHOCHO and HCHO from OMI with ground-based MAX-DOAS observations of those molecules over Mexico City and the Pearl River Delta, and with global simulations from the GEOS-Chem chemical transport model, for the study of VOCs for air quality. The seasonally-resolved variations of simultaneous, space-based observations of formaldehyde and glyoxal allow for the first-ever study of this kind.

While HCHO has been observed from space-based instruments for some time now (Chance *et al.*, 2000), the first ground-based measurements of CHOCHO were reported only recently (Volkamer *et al.*, 2005), followed shortly by the first space-based observations of this molecule (Kurosu *et al.*, 2005; Wittrock *et al.*, 2006). Glyoxal is produced from the oxidation of a wide range of VOCs, including isoprene, alkene, alkyne, and aromatics. Like HCHO, its major gas-phase sinks are photolysis and the reaction with OH. Unlike HCHO, glyoxal is virtually unaffected by vehicle emissions and is therefore a better indicator for the reactivity of the urban atmosphere (photochemical smog)) (Volkamer *et al.*, 2005). Recent laboratory studies have also suggested heterogeneous aerosol uptake as a significant sink of glyoxal (Liggio *et al.*, 2005).

Due to the as-yet limited number of observations, three major outstanding questions relating to glyoxal are: 1. its global distribution and budget; 2. the ratio of CHOCHO column amounts to those of NO_x and formaldehyde; and 3. its seasonal variation. The present study addresses all three questions.

The work is currently in progress but will be completed well before the SBSR conference. At the time of this writing, the status of the individual components is as follows:

- The processing of 12 months of CHOCHO (11/2005-10/2006) from OMI is complete; the extension of the time series is ongoing.
- HCHO processing from OMI (both forward and backward in time) is ongoing and currently covers 04/2006 to mid-11/2006.
- MAX-DOAS measurements of CHOCHO and HCHO have been performed in the Pearl River Delta and are currently being evaluated.
- Global simulations of CHOCHO and HCHO with GEOS-Chem are under way.

2. The Ozone Monitoring Instrument

The Ozone Monitoring Instrument (OMI) was launched on-board EOS Aura on 15 July 2004 into a sun-synchronous orbit with an equator crossing time of 1338h (ascending node). OMI is a UV/Vis CCD spectrometer that covers the spectral range of 270-500 nm with a spectral resolution of 0.45-1.0 nm FWHM. A total across-track swath of 2600 km, divided into 60 pixels of sizes ranging from 14×24 km^2 at nadir to about 28×150 km^2 at the edges of the swath, provides daily global coverage. OMI routinely observes ozone, aerosol and cloud products, NO_2, SO_2, BrO, OClO, and HCHO. It is the first instrument from which space-based observations of glyoxal are reported (Kurosu *et al.*, 2005).

3. Retrieval Algorithm and Air Mass Factor Scheme

The retrievals of HCHO and CHOCHO are performed in the spectral windows of 325-357 nm and 421-458 nm respectively. The retrieval procedure determines slant column abundances Ω_s by non-linear least-squares fitting of measured radiances and irradiances, including molecular absorption cross sections, correction for Ring effect, albedo, and a low-order closure polynomial (Kurosu *et al.*, 2004). The Ω_s are then converted to vertical columns Ω_v by ways

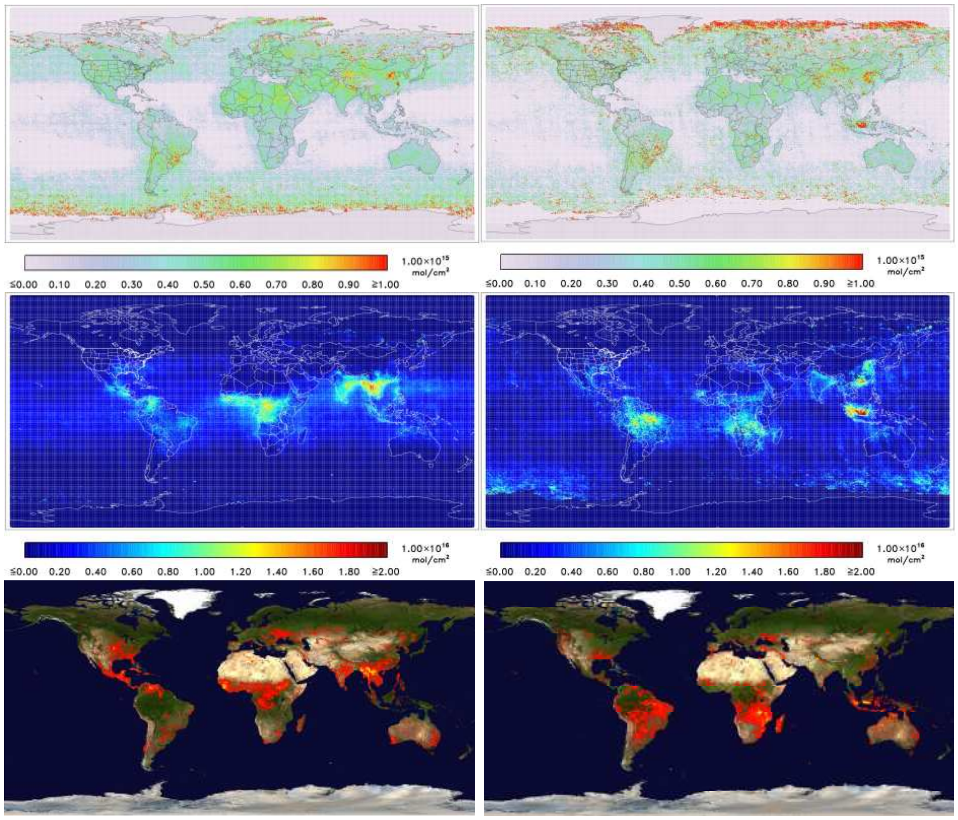

Figure 1: OMI CHOCHO (top) and HCHO (middle) vertical columns with MODIS fire counts (bottom) for the two 10-day time periods 1-10 April 2006 (left column) and 8-17 October 2006 (right column). While HCHO is strongly correlated with biomass burning, CHOCHO columns are elevated only over most intense burning events but are correlated more strongly with anthropogenic sources. Fire counts have been obtained from MODIS Rapid Response System Global Fire Maps at <http://rapidfire.sc.gsfc.nasa.gov/firemaps/>.

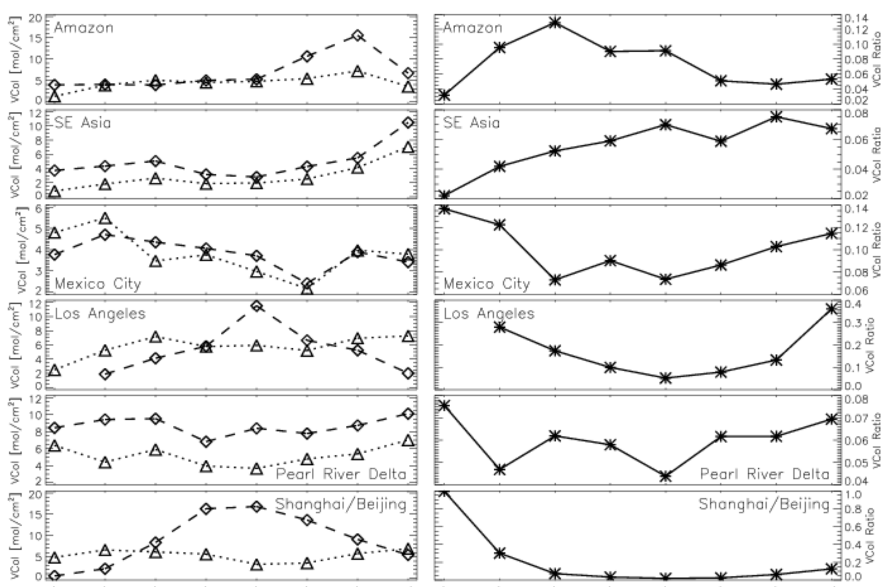

Figure 2: (left) Time series of OMI vertical column HCHO (diamonds) and CHOCHO (triangles) monthly averages for March-October 2006, over selected geographical areas with predominantly anthropogenic or biogenic VOC emissions; column values have been normalized to 10^{15} (HCHO) and 10^{14} (CHOCHO) mol/cm^2 respectively. (right) CHOCHO-to-HCHO ratio of vertical columns from the left panels. Typical values range between 0.04 to 0.1 and vary more strongly by region than by VOC type.

of a molecule-specific air mass factor, AMF = Ω_s/Ω_v. This AMF contains information on the vertical distribution of the target gas, and includes scattering weights to take into account the altitude-dependent sensitivity of the nadir-viewing satellite measurements. While AMF tables have been computed for HCHO, no AMFs are available for glyoxal, mainly due to the still sparse knowledge of its global distribution. However, to 0^{th} order approximation, CHOCHO spatial and vertical distribution follows that of HCHO, hence the HCHO AMFs, scaled with albedo- and viewing-geometry dependent correction factors based on the difference in vertical sensitivity of the HCHO and CHOCHO observations, have been used to derive CHOCHO vertical columns. This AMF scheme will be improved as soon as GEOS-Chem simulations of horizontal and vertical distributions of glyoxal are available.

4. OMI Observations of CHOCHO and HCHO

Figure 1 shows CHOCHO (top panels) and HCHO (middle) vertical columns for the two 10-day periods 1-10 April 2006 (left column) and 8-17 October 2006 (right column). While HCHO is strongly correlated with biomass burning, CHOCHO columns are elevated only over the most intense burning events but are also correlated more strongly with anthropogenic sources: note the enhancements over Mexico City, Turin, the Ruhr valley in Germany, Riyadh, Guangzhou, Shanghai, Beijing, Tokyo, and other cities. Additional global images of CHOCHO and HCHO for other time periods can be found at
<http://www.cfa.harvard.edu/~tkurosu/SatelliteInstruments/OMI/SampleImages/index.html>

Figure 2 contains time series of CHOCHO and HCHO monthly average columns from March to October 2006 for selected areas of either megacities (Los Angeles, Mexico City, the Pearl River Delta close to Hong Kong, Shanghai/Beijing) or biomass burning (the Amazon, South-East Asia). In the panels on the left, CHOCHO (triangles) and HCHO (diamonds) vertical column values are shown, normalized to 10^{14} and 10^{15} respectively. The panels on the right show the corresponding CHOCHO-to-HCHO ratios. Typical values for these ratios range between 0.04 and 0.1, depending on season, and varying strongly by region rather than by the nature of the emission sources (anthropogenic/biogenic/biomass burning). This indicates that the CHOCHO/HCHO ratio is primarily controlled by the particulars of VOC speciation in each region. Further studies of the spatial and temporal variation of this novel OVOC ratio need to be performed on a case-by-case basis.

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