Anthropogenic emissions of highly reactive volatile organic compounds inferred from oversampling of OMI HCHO columns

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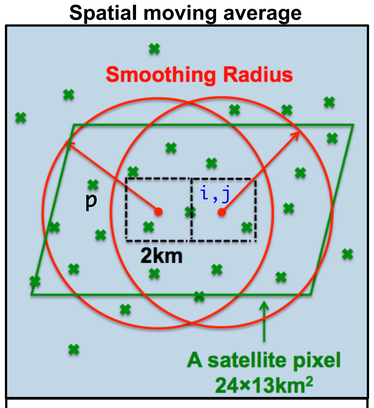
1. Abstract

Satellite observations of formaldehyde (HCHO) columns provide top-down constraints on emissions of highly reactive volatile organic compounds (HRVOCs). This approach has been used previously in the US to estimate isoprene emissions from vegetation, but application to anthropogenic emissions has been stymied by lack of a discernable HCHO signal. Here we show that temporal oversampling of HCHO data from the Ozone Monitoring Instrument (OMI) for 2005–2008 enables detection of urban and industrial plumes in eastern Texas including Houston, Port Arthur, and Dallas/Fort Worth. By spatially integrating the HCHO enhancement in the Houston plume observed by OMI we estimate an anthropogenic HCHO source of 250 ± 140 kmol h^{-1}. This implies that anthropogenic HRVOC emissions in Houston are 4.8 ± 2.7 times higher than reported by the US Environmental Protection Agency (EPA) inventory, and is consistent with field studies identifying large ethene and propene emissions from petrochemical industrial sources. This approach allows us to identify potential anthropogenic HCHO hot spots in China, and to monitor trends in HRVOC emissions over the US, in particular from the urban areas and the oil/gas industry.

2. Data and methods

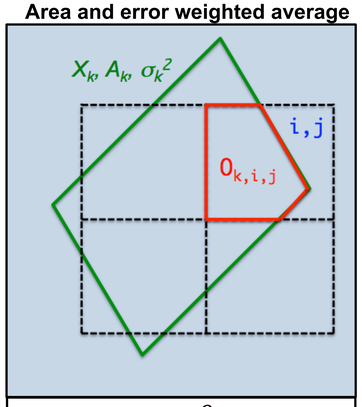
- OMI level-2 data (**version 2 and 3**): Harvard-Smithsonian Center for Astrophysics
- Surface albedo: OMI Surface Reflectance Climatology Data Product-OMLER
- 13:00–14:00 local time temperatures: MERRA
- Air mass factors (AMF) are calculated by a nested GEOS-Chem (0.5×0.667°) model and a radiative transfer model, LIDORT. Slant column densities (SCD) are converted to vertical column densities (VCD, column hereafter) using AMF.
- Bottom-up AHRVOC emissions are from US EPA NEI05 as implemented by Stuart McKeen [Brioude *et al.* 2011, Kim *et al.* 2011].
- Drift from instrument aging is removed with a linear temporal regression of background SCD over the North Pacific.
- Two oversampling methods:

Method 1
Spatial moving average



$$\overline{X_{i,j}} = \{X_p, where\ p\ within\ the\ circle\}$$

Method 2
Area and error weighted average



$$\overline{X_{i,j}} = \frac{\sum_{k=1}^{N_{i,j}} \frac{O_{k,i,j}}{A_k \sigma_k^2} X_k}{\sum_{k=1}^{N_{i,j}} \frac{O_{k,i,j}}{A_k \sigma_k^2}}$$

References

- Brioude J, *et al.*, [2011], Top-down estimate of anthropogenic emission inventories and their interannual variability in Houston using a mesoscale inverse modeling technique, *J. Geophys. Res.* 116 D20305
- Kim S-W, *et al.*, [2011], Evaluations of NOx and highly reactive VOC emission inventories in Texas and their implications for ozone plume simulations during the Texas air quality study 2006, *Atmos. Chem. Phys.* 11 11361–86
- Zhu L, et al., [2014], Anthropogenic emissions of highly reactive volatile organic compounds in eastern Texas inferred from oversampling of satellite (OMI) measurements of HCHO columns, *Environ. Res. Lett.* 9 114004

3. HCHO columns in east Texas

OMI HCHO columns (**version 2**) over May–August 2005–2008 were oversampled to a 0.02° x 0.02° resolution using an averaging radius of 24 km (**Method 1**).

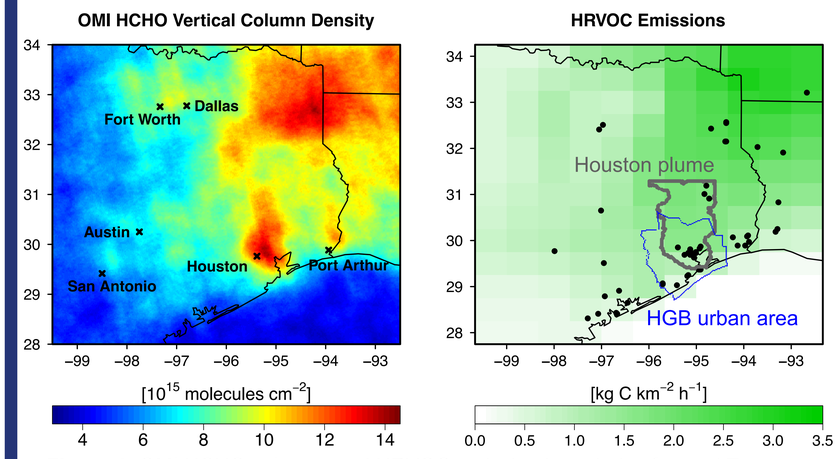


Figure 1. OMI HCHO columns and HRVOC emission inventories for east Texas. The right panel shows distribution of May–Aug. 2008 MEGAN isoprene emissions (background) and NEI05 major AHRVOCs point sources with emissions larger than 3 kg C h^{-1} (dots). See **Figure 5** for emission breakdown.

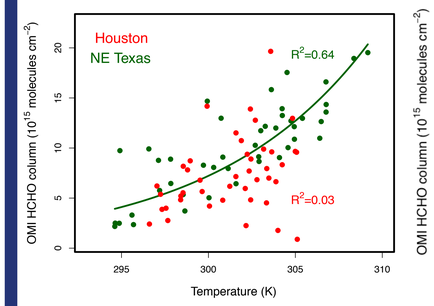


Figure 2. Relationship of OMI HCHO column with surface air temperature for the Houston urban core and Northeast Texas. Individual points are ten-day averages for May–Sep., 2006–2008.

One can use temperature to distinguish biogenic and anthropogenic HCHO.

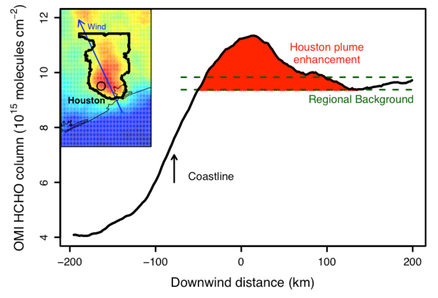


Figure 3. Cross-section of the Houston HCHO plume along the direction of the prevailing SSE wind. This shows the mean HCHO columns for May–Aug. 2005–2008 averaged across the plume width as a function of downwind distance from Houston.

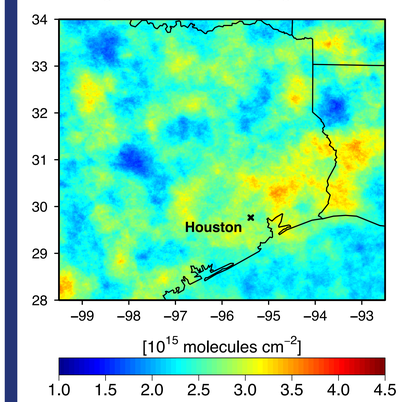


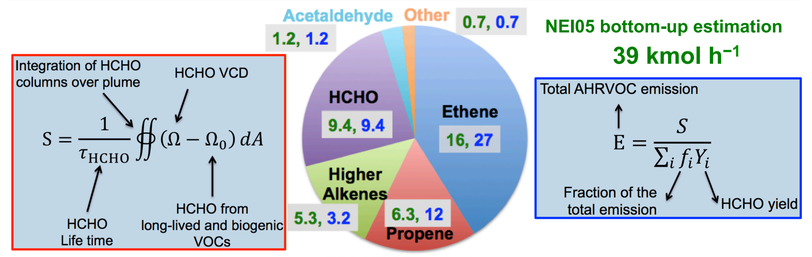
Figure 4. 2005-2008 winter time (DJF) oversampled HCHO columns.

The lack of detectable OMI HCHO enhancements in winter is mainly due to lower OH and higher wind speed in winter.

This suggests that anthropogenic HCHO is mainly produced by photochemical oxidation of alkenes (secondary) rather than directly emitted (primary).

For more about section 3 and 4, see Zhu *et al.* [2014]

4. Inferring AHRVOC emissions using HCHO columns



Anthropogenic enhancement
250 ± 140 kmol HCHO h^{-1}

Top-down constraint
190 ± 100 kmol h^{-1}

Figure 5. Major AHRVOC emissions and related HCHO production rates in Houston plume

AHRVOC emissions in Houston are underestimated by a factor of **4.8 ± 2.7** by EPA.

5. Interannual variability in HCHO columns over east Texas

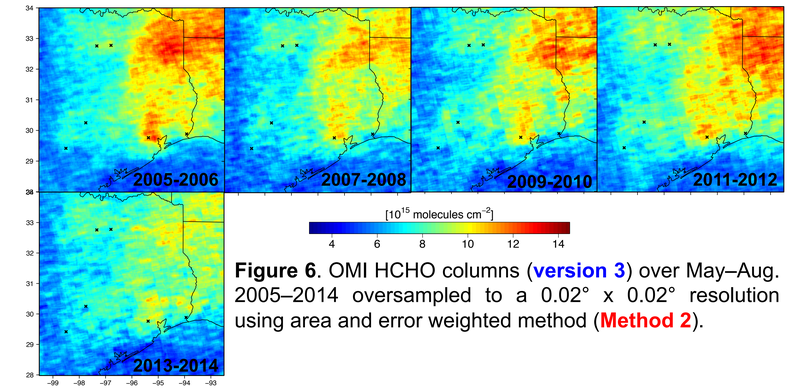


Figure 6. OMI HCHO columns (**version 3**) over May–Aug. 2005–2014 oversampled to a 0.02° x 0.02° resolution using area and error weighted method (**Method 2**).

HCHO columns in east Texas show large interannual variability in the past decade. One need to do temperature correction for biogenic HCHO in order to get HCHO trend from AHRVOCs.

6. Our method can detect HCHO hot spots in China

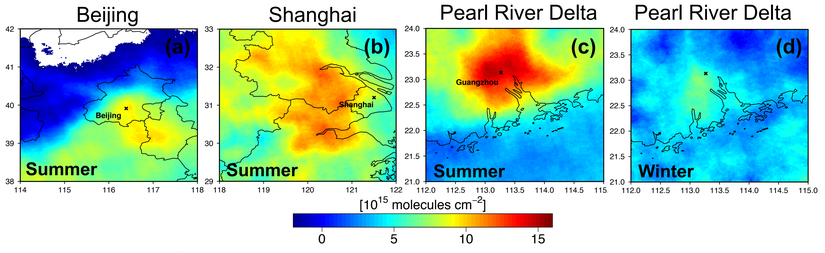


Figure 7. OMI HCHO oversampling results (**version 2, Method 1,** 0.02° x 0.02°) during 2005–2008 summer (MJJAS, a–c) and winter time (DJF, d) for different urban areas in China.

Acronyms:
(A)HRVOCs: (Anthropogenic) Highly Reactive Volatile Organic Compounds
AMF: Air Mass Factor;
HGB: Houston–Galveston–Brazoria urban metropolitan area
LIDORT: LInearized Discrete Ordinate Radiative Transfer
MEGAN: The Model of Emissions of Gases and Aerosols from Nature
MERRA: The Modern-Era Retrospective analysis for Research and Applications
OMI: The Ozone Monitoring Instrument; **SCD**: Slant Column Density; **VCD**: Vertical Column Density

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