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

OMI HCHO 2006 JJA average

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Relating HCHO columns to HRVOC emissions

\[ \text{Emission } E_i \]

\[ \text{VOC}_i \xrightarrow{\text{oxidation}} \text{HCHO} \xrightarrow{h\nu (340 \text{ nm}), \text{OH}} \]

\[ k \approx 0.5 \text{ h}^{-1} \]

In absence of horizontal wind, mass balance for HCHO column \( \Omega_{\text{HCHO}} \):

\[ \Omega_{\text{HCHO}} = \sum_i y_i E_i \]

but wind smears this relationship

Detection Limit

VOCs source

Distance downwind

Isoprene

Ethene

Propene

Methanol
Oversampling approach to detect point/urban sources

- Oversampling: temporal averaging of the satellite data on a spatial grid finer than the pixel resolution of the instrument.
- Takes advantage of the spatial offset and changing geometry of the satellite pixels from day to day.
- Trades temporal for spatial resolution.
- Achieves higher signal-to-noise ratio.

Optimize smoothing radius:
- Too fine (12 km): Increase noise.
- Too coarse (36 km): Lose spatial features.

A satellite pixel

24×13 km²

2005-2008 MJJAS

26028 pixels in a 1°×1° box
Results: Oversampling of OMI HCHO pixels

OMI HCHO column, 2005-2008, MJJA

OMI HCHO column

(10^{15} \text{ molecules cm}^{-2})

Temperature (K)

Houston

NE Texas

R^2=0.64

R^2=0.03

Oversampling approach enables detection of anthropogenic HRVOCs from urban/industrial sources and oil/gas operations.
Deriving the HCHO source in the Houston plume

Integration of HCHO columns over plume

\[ S = \frac{1}{\tau_{\text{HCHO}}} \int \int (\Omega - \Omega_0) \, dA \]

HCHO VCD
HCHO from long-lived and biogenic VOCs
Regional Background
Anthropogenic Enhancement

Houston
Coastline

OMI HCHO column (10^{15} \, \text{molecules cm}^{-2})

Downwind distance (km)
Inference of AHRVOC emissions from the HCHO columns

\[ S = \frac{1}{\tau_{\text{HCHO}}} \int \int (\Omega - \Omega_0) \, dA \]

HCHO lifetime: 1.6±0.5 h

\( S: 250\pm140 \text{ kmol HCHO h}^{-1} \)

Bottom-up estimate:
\( 240\pm90 \text{ kmol HCHO h}^{-1} \) [Parrish et al., 2012]

Total AHRVOC emission

\[ E = \frac{S}{\sum_i f_i Y_i} \]

Fraction of the total emission

HCHO yield

AHRVOC emissions in the Houston plume area are underestimated by a factor of 4.8±2.7 in EPA NEI05 inventory for 2005–2008.
Indistinguishable HCHO enhancements in winter at Houston

OMI HCHO 2005–2008 MJJA

OMI HCHO 2005–2008 DJF

HCHO enhancements at Houston are indistinguishable during winter even by oversampling, due to smearing resulting from low OH and high wind speed.

This suggests that anthropogenic HCHO is mainly secondary rather than primary.
Oversampling of OMI HCHO pixels in China, 2005-2008

Aoxing Zhang and Lei Zhu
Future work

• Improving the oversampling technique: e.g., using Gaussian or inverse distance weights for spatial smoothing.

• Detecting long-term trends of HCHO in urban/industrial areas and oil/gas fields.

• Looking at HCHO over China.

• Linking HCHO with other information, e.g., wind speed, wind direction, or glyoxal columns.

Take home messages

• Oversampling of OMI HCHO columns solves the long-standing problem of detecting and quantifying US AHRVOC emissions from space.

• AHRVOC emissions for Houston are $4.8 \pm 2.7$ times higher than that in EPA inventory.

• Due to low OH and high wind speed, OMI HCHO enhancements in winter are indistinguishable at Houston, which suggests that anthropogenic HCHO is mainly secondary.