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Joint inversion of formaldehyde and isoprene to constrain global non-methane VOC emissions

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INTRODUCTION & OBJECTIVES

Emissions of non-methane volatile organic compounds (VOCs) have large uncertainties owing to the diversity of VOC species and sources, and lack of up-to-date emission factors and reliable activity statistics.

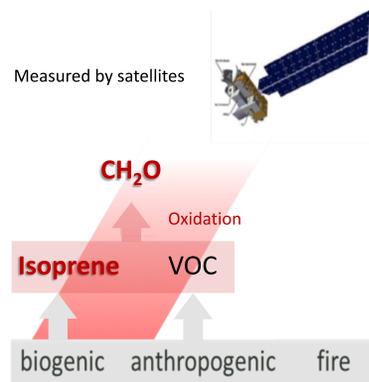
CH₂O is a high-yield intermediate product of the oxidation of VOCs. Space based observations of CH₂O have been widely used as a proxy for VOC emissions.

Constraining VOC emissions using CH₂O observations alone is limited because of lack of species- and sector-specific information (Choi et al., 2022).

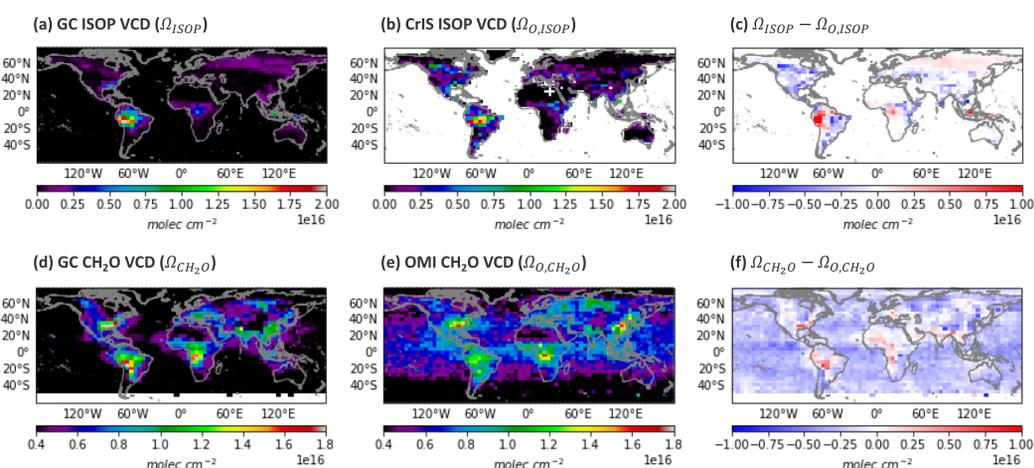
Isoprene is the dominant VOC emitted to the global atmosphere. The emissions are almost entirely biogenic. Therefore, information from isoprene observations could help quantifying VOC emission speciation and sectoral contribution.

We seek to improve our understanding of global VOCs emissions using the OMI CH₂O and the CrIS isoprene retrievals (Wells et al., 2020), and to better understand the sources of O₃ and PM_{2.5}.

As a preliminary study, we explore global VOC emissions in July 2016.



OMI CH₂O & CrIS ISOPRENE RETRIEVALS (JULY 2016)

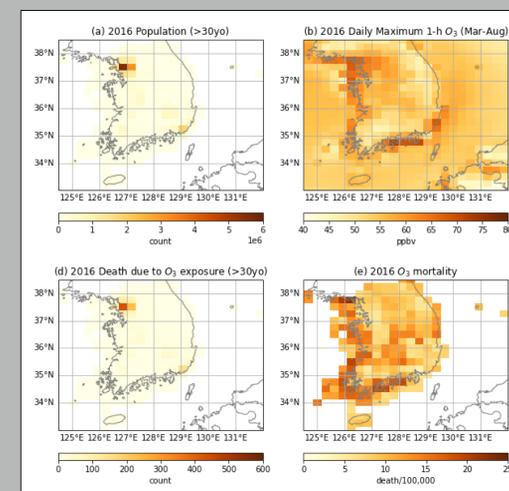


Vertical column densities (VCD) of isoprene (upper row) and CH₂O (lower row) of the a priori GEOS-Chem simulation (left column) and satellites (middle column). Differences between the simulation and the observations (right column).

CONTRIBUTIONS OF LOCAL SOURCES ON O₃ RELATED PREMATURE DEATHS IN KOREA

We aim to quantify the contribution of VOC emissions in different source regions to O₃ related health burden in Korea. We use GEOS-Chem v35n in 0.25°×0.3125° with the optimized VOC emissions (Choi et al., 2022).

2,731 premature deaths owing to O₃ exposure in 2016 are estimated



Population and Mortality Data

from Korean National Statistics 2016

Simulated surface O₃ concentrations

from GEOS-Chem forward model

Mortality attributable to O₃ exposure

Jerrett et al., 2009

O₃-attributable premature deaths (D_{O_3})

$$D_{O_3} = D \exp\left(1 - HR \frac{F(O_3) - TMREL}{\Delta O_3}\right)$$

D is the deaths from a disease

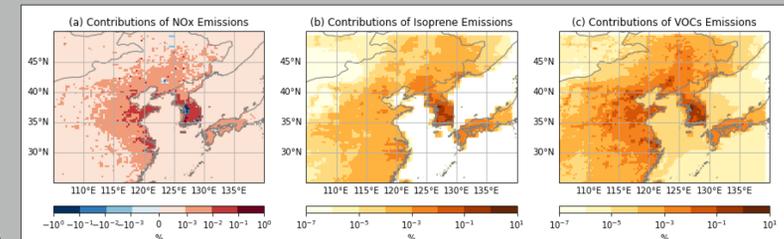
HR is the hazard ratio

$F(O_3)$ is the 6-mon daily maximum 1-h O₃

$TMREL$ is the minimum O₃ level cutoff

ΔO_3 is the O₃ incremental changes to which the mortality is attributable

Effectiveness of domestic emission control is 25% (that of Chinese emission control is 31%)



Effectiveness of incremental control of emissions in reducing O₃ related premature deaths in 2016

Model Configuration	
GEOS-Chem	13.3.3
MERRA2	4°×5° 72L
Default global emission inventories	
CEDSv2	
MEGANv2.1 (OFFLINE_BIOGENICVOC)	
GFED4	
VOC emission species	
ACET, ALD2, ALK4, BENZ, C2H2, C2H4, C2H6, C3H8, CH2O, EOH, GLYC, HAC, HCOOH, ISOP, LIMO, MACR, MEK, MOH, MTPA, MTPO, MVK, PRPE, RCHO, TOLU, XYLE	

In this study, we define the vector of VOC emissions (\mathbf{E}), and the vector of vertical column densities (VCDs; Ω) at each model grid cell following Qu et al. (2019).

$$\mathbf{E} = \begin{bmatrix} E_{ISOP} \\ E_{VOC} \end{bmatrix}, \quad \Omega = \begin{bmatrix} \Omega_{ISOP} \\ \Omega_{CH_2O} \end{bmatrix}$$

where E_{ISOP} is the isoprene emissions, E_{VOC} is the total non-isoprene VOC emissions, and

Ω_{ISOP} and Ω_{CH_2O} are the VCD of isoprene and CH₂O, respectively

A first order Taylor expression of Ω around the a posteriori VOC emissions (\mathbf{E}_a) is

$$\Omega = \Omega_a + \mathbf{H}(\mathbf{E} - \mathbf{E}_a).$$

where \mathbf{H} is the Jacobian matrix, which is defined as

$$\mathbf{H} = \begin{bmatrix} \frac{\partial \Omega_{ISOP}}{\partial E_{ISOP}} & \frac{\partial \Omega_{ISOP}}{\partial E_{VOC}} \\ \frac{\partial \Omega_{CH_2O}}{\partial E_{ISOP}} & \frac{\partial \Omega_{CH_2O}}{\partial E_{VOC}} \end{bmatrix}$$

JOINT MASS BALANCE INVERSION USING CH₂O AND ISOPRENE MEASUREMENTS

Cost function of the joint inversion

We optimize global total VOC emissions using a joint mass balance framework. The method aims to minimize the cost function (J), which is defined at each grid cell as

$$J = J_o + J_a$$

$$= \frac{1}{2}(\Omega - \Omega_{obs})^T \mathbf{R}^{-1}(\Omega - \Omega_{obs}) + \frac{1}{2}\gamma(\mathbf{E} - \mathbf{E}_a)^T \mathbf{B}^{-1}(\mathbf{E} - \mathbf{E}_a)$$

$$= \frac{1}{2}(\Omega_a + \mathbf{H}(\mathbf{E} - \mathbf{E}_a) - \Omega_{obs})^T \mathbf{R}^{-1}(\Omega_a + \mathbf{H}(\mathbf{E} - \mathbf{E}_a) - \Omega_{obs})$$

$$+ \frac{1}{2}\gamma(\mathbf{E} - \mathbf{E}_a)^T \mathbf{B}^{-1}(\mathbf{E} - \mathbf{E}_a),$$

where J_o is the error-weighted squared difference between Ω and Ω_a , and J_a is the error-weighted squared difference between \mathbf{E} and \mathbf{E}_a , Ω_{obs} is the VCDs measured by OMI and CrIS, \mathbf{R} is the observation error covariance matrix, \mathbf{B} is the prior error covariance matrix, and γ is the regularization parameter for J_a . We use $\gamma = 1.0$ for the preliminary study.

Solution of the joint inversion

The posteriori emission ($\hat{\mathbf{E}}$) is the solution of $\frac{\partial J}{\partial \mathbf{E}} = 0$.

$$\hat{\mathbf{E}} = \mathbf{E}_a + (\mathbf{H}^T \mathbf{R}^{-1} \mathbf{H} + \mathbf{B}^{-1})^{-1} \mathbf{H}^T \mathbf{R}^{-1} (\Omega_{obs} - \Omega_a)$$

Covariance matrices

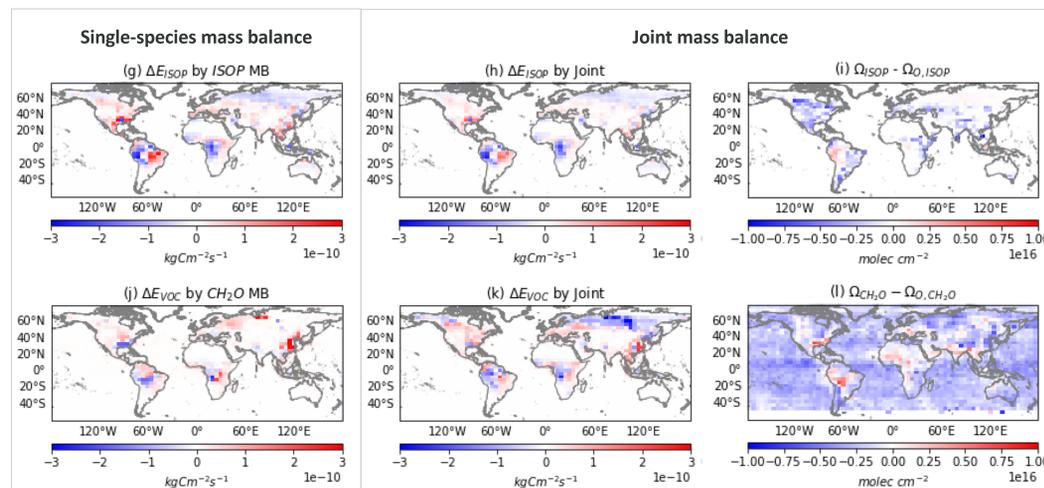
We estimate the covariance matrices to be diagonal.

The diagonal of \mathbf{R} is the variance of Ω_a . \mathbf{B} is defined as

$$\mathbf{B} = \begin{bmatrix} \sigma_{a,ISOP}^2 / \gamma_{ISOP}^{MB} & 0 \\ 0 & \sigma_{a,VOC}^2 / \gamma_{VOC}^{MB} \end{bmatrix}$$

where $\sigma_{a,x}$ is the a priori uncertainty of emission species x , and γ_x^{MB} is the regularization parameter calculated in a single-species mass balance inversion. γ_{ISOP}^{MB} is 1.0 as a proxy, and γ_{VOC}^{MB} is 0.1 following Choi et al. (2022).

COMPARISONS OF TOP-DOWN VOC EMISSIONS BETWEEN JOINT & SINGLE-SPECIES MASS BALANCE



Differences between the a posteriori and the a priori emissions of isoprene (upper row) and non-isoprene VOC (lower row) when assimilating isoprene and CH₂O observations individually (left column) and jointly (middle column). Differences between the joint a posteriori simulation and the observations (right column) of the vertical column densities of (upper row) isoprene and CH₂O (lower row). The VOC emissions constrained by the CH₂O mass balance inversion include the isoprene emissions, whereas those constrained by the joint mass balance inversion are non-isoprene.

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ISSUE & FUTURE WORK

- Validation:** Evaluate with ground and aircraft observations of VOCs and O₃
- Emission reduction is calculated stronger than emission increase. Sensitivities of non-isoprene VOC emissions to isoprene VCD leads to emission reduction in Asia:** (i) Constrain absolute emissions or log of emission scaling factor. (ii) Conduct a sensitivity analysis to optimize the regularization parameters
- Nonlinearity:** (i) Perform joint inversion iteratively. (ii) Understand the roles of Jacobian matrix, \mathbf{H} . (iii) Explore impacts of VOC emissions to atmospheric oxidation, O₃ and PM_{2.5}.
- Constrain VOC emissions for other seasons and years (2013–2017) in a higher resolution (2°×2.5°).