Interannual variability and trend analysis of atmospheric formaldehyde in boreal autumn over recent decades

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1. Summary

This study examines the spatial-temporal variations of atmospheric CH_2O and investigates regional sources influencing its interannual variability (IAV) and trends over the past two decades. Using recent data retrievals from the Ozone Monitoring Instrument (OMI) on NASA's Aura satellite, simulations from the NASA Goddard Earth Observing System Chemistry Climate Model (GEOSCCM) and a multiple linear regression model (MLR), we assess the impact of anthropogenic volatile organic compounds (VOCs) emissions, biogenic emissions, open fires and climatic variations on CH_2O IAV and trends.

Key findings:

RefD1 reproduces the observed spatial variations in CH_2O, but with notable overestimations in southeast US, South America and southern Africa.

For IAV:

i. RefD1 reproduces the observed IAV over eastern China and South America.

ii. Isoprene plays an important role in controlling the IAV of CH_2O over most regions, except in Indonesia, where VOCBB (VOCs from biomass burning) dominates and correlates with the El Niño-Southern Oscillation (ENSO).

iii. VOCBB is another crucial factor in three biomass burning regions.

iv. ENSO shows a minor negative impact on IAV over Northern Africa and South America.

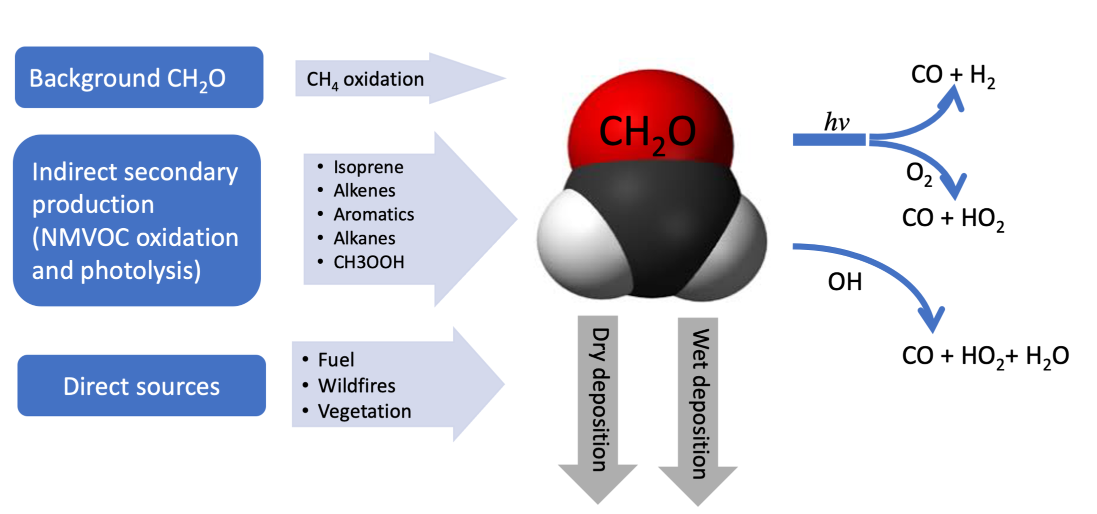
For trends:

I. VOCFF (VOCs from fossil fuel emissions) significantly impact CH_2O trend over Eastern China, India, Northern Africa and Indonesia.

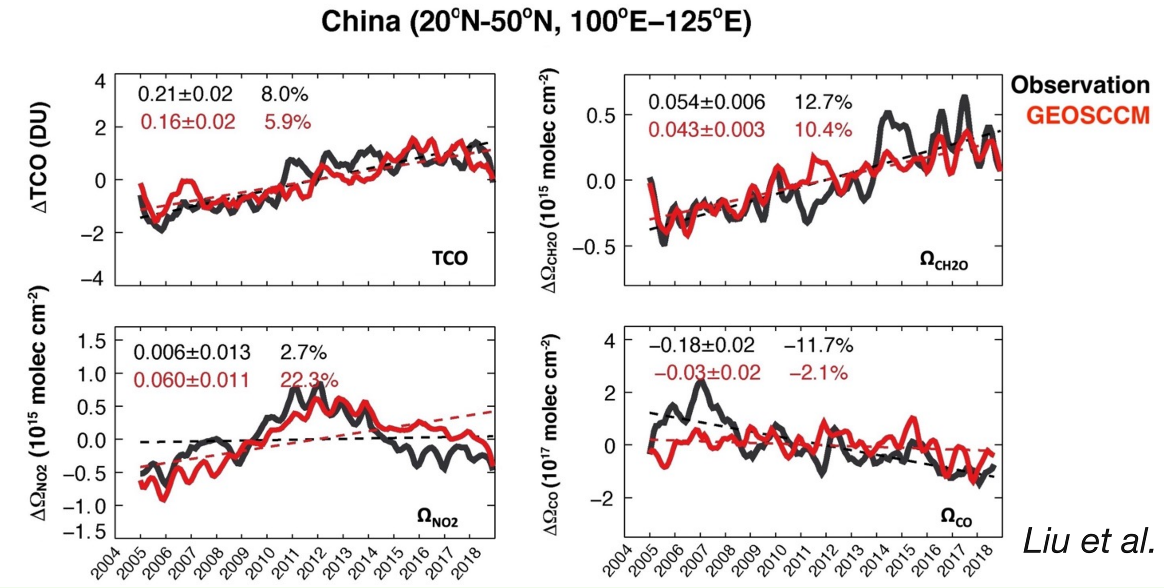
II. Isoprene drives positive trends in simulated CH_2O over South America, despite a negative VOCBB trend in the region.

III. Sources other than isoprene, VOC emissions, and ENSO impact simulated trends in the Southeast US, India and Northern Africa.

2. Introduction



- **CH_2O: an intermediate product gas in the oxidation chains of VOCs, with a short atmospheric lifetime of a few hours.**
- **A reliable localized proxy of VOCs, given that most VOCs cannot be directly observed from space.**



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The notable increase of TCO over China is likely driven by the increase of VOCs, reflected by the positive trend in CH_2O.

Key questions:

- **What are the spatial-temporal variations of atmospheric CH_2O?**
- **What are the key factors influencing interannual variations and trends within identified CH_2O hotspots?**

3. Data and Model

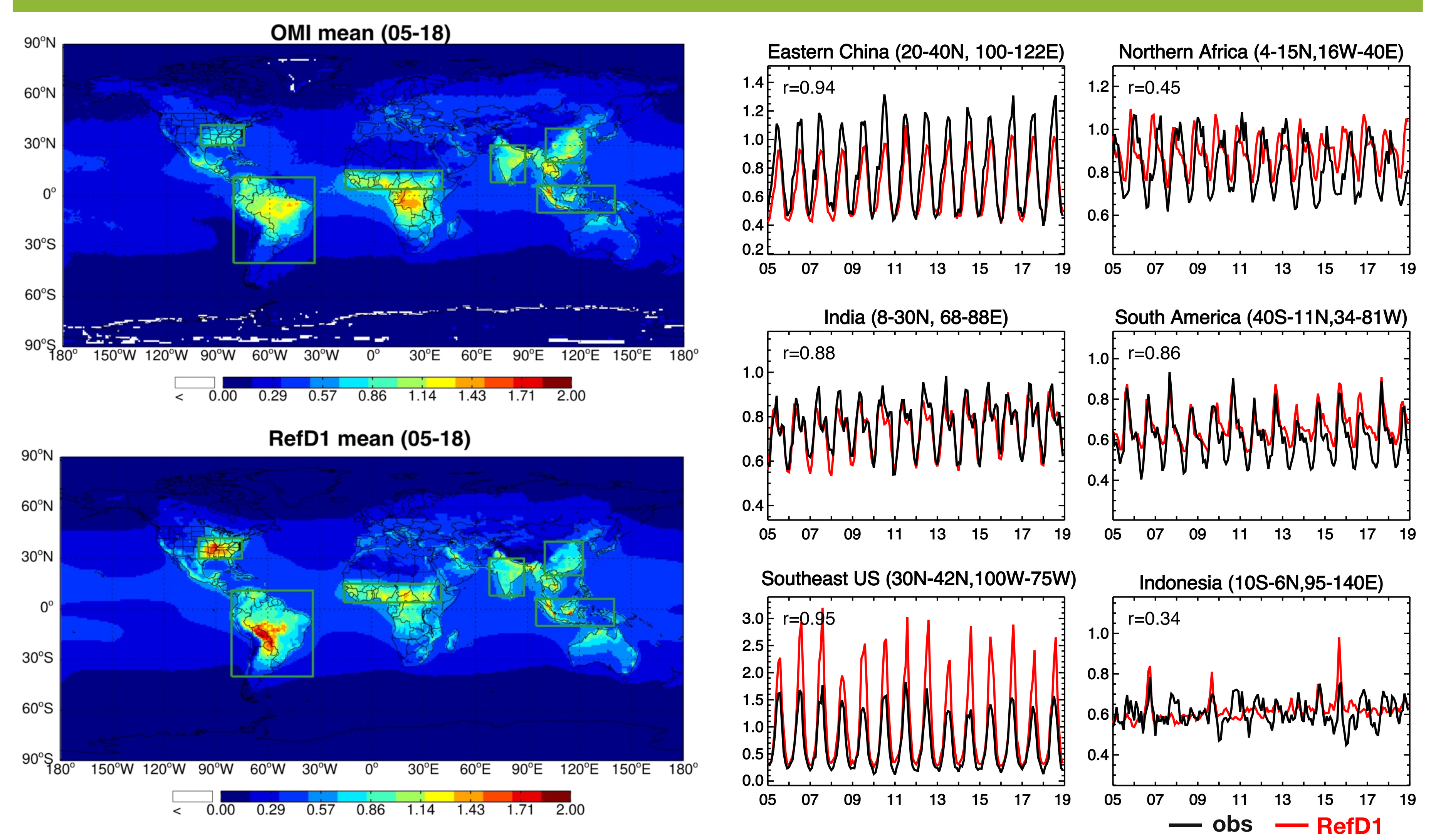
Satellite observations - OMI Smithsonian Astrophysics Observatory (SAO) CH_2O retrieval V4:

- updated Level 1B radiances
- an improved radiative transfer model
- a new reference sector correction methodology
- Better agreement with OMPS (Ozone Mapping and Profiler Suite) CH_2O retrieval

GEOSCCM RefD1 simulation:

- Free running hindcast simulation of 1960-2018 supporting the Chemistry-Climate Modeling Initiative (CCMI)
- GMI chemistry, GOCART aerosol and an artificial stratospheric O_3 tracer ($StatO_3$).
- C90 resolution, 72 vertical levels
- Anthropogenic emissions are from CEDS (1980–2014) and SSP2–4.5 (2015-2017)
- Biomass burning emissions are from CMIP6 harmonized emission inventory (1980-2015) and GFED4 (2016-2017)
- Biogenic emissions through online calculation with Model of Emissions of Gases and Aerosols from Nature (MEGAN).

4. Initial model evaluation



- **RefD1 reproduces the observed spatial variations in CH_2O, showing**
  - **Locally elevated mixing ratio over the southeast US, South America, equatorial and southern Africa, India, Eastern China, Indonesia.**
  - **Low CH_2O mixing ratio over the remote oceanic region, mainly from methane oxidation.**
- **RefD1 shows notable overestimations in southeast US and South America, along with underestimations in southern Africa.**
- **Model reproduces seasonal cycles of observed CH_2O over most regions.**

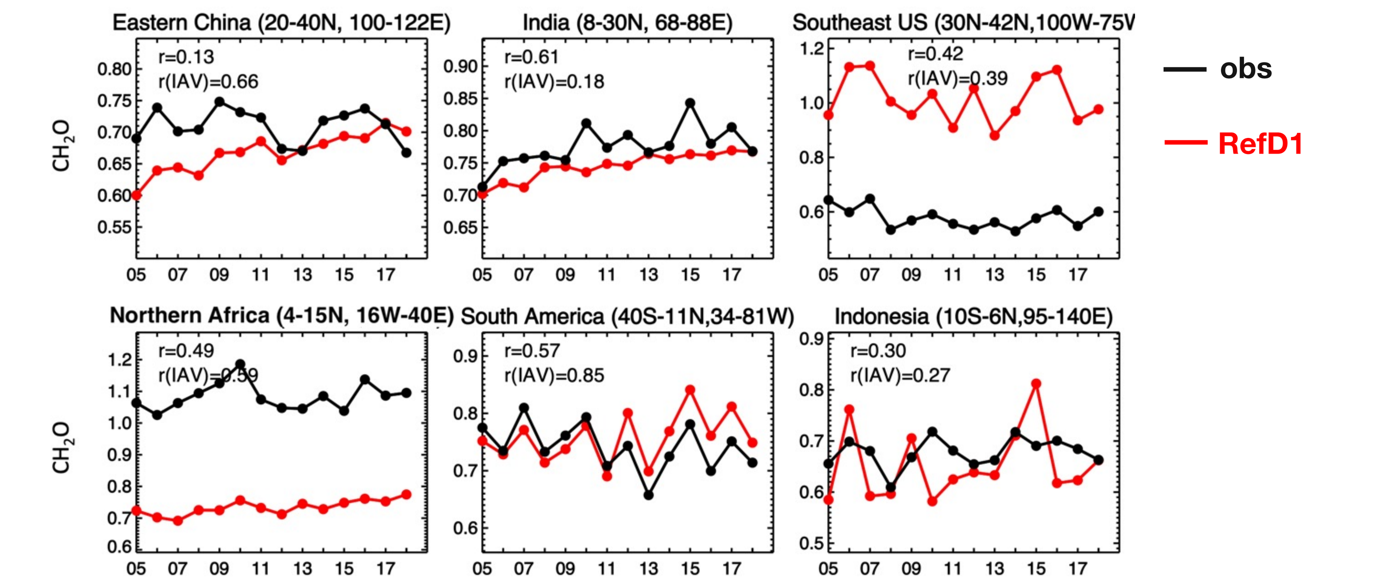
5. Sources attributions - Regional Multiple Linear Regression (MLR) Analysis

$$CH_2O(t) = \alpha + \beta_0 t + \beta_1 * ISOP(t) + \beta_2 * VOCBB(t) + \beta_3 * VOCFF(t) + \beta_4 * ENSO(t) + R(t)$$

- $CH_2O(t)$: Regional area weighted CH_2O
- $ISOP(t)$: Isoprene biogenic source
- $VOCBB(t)$: VOC emissions from biomass burning
- $VOCFF(t)$: VOC emissions from anthropogenic source
- $ENSO(t)$: Nino 3.4

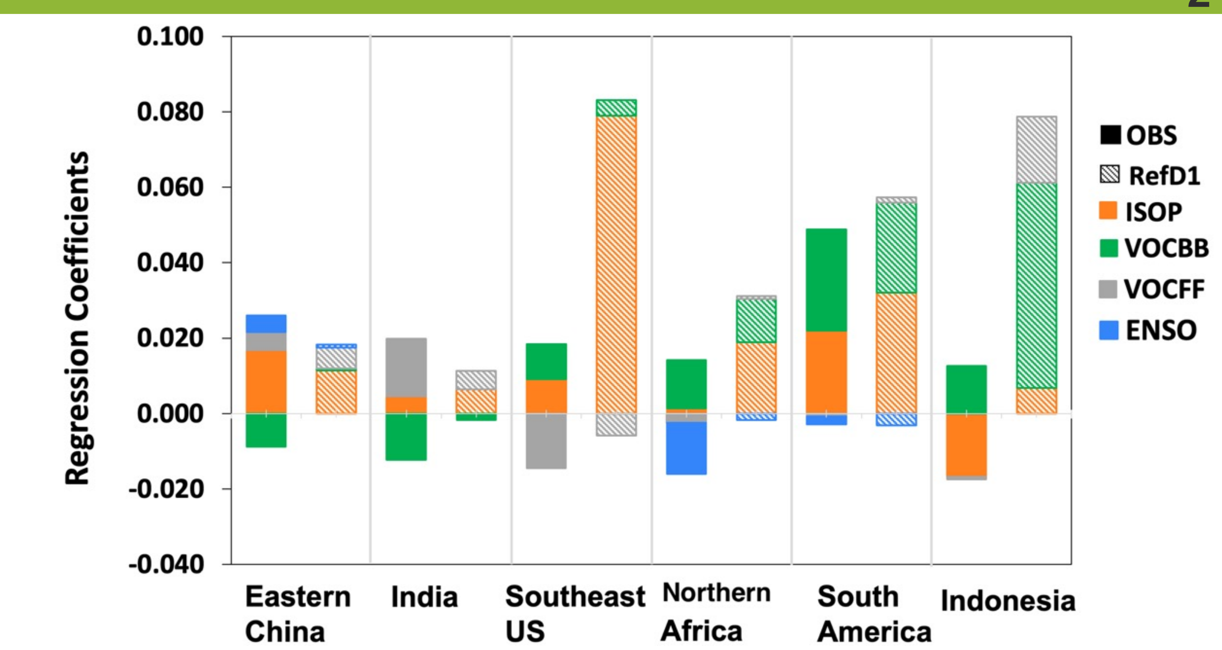
- **MLR analysis from 2005 to 2018 during boreal autumn (SON) over six selected regions.**
- **VOCs: The sum of CH_2O, C_4H_8O, C_3H_6, and higher alkenes, C_2H_6, C_3H_8, C_4H_{10}, C_2H_4O.**
- **Excluding TS in the MLR due to its high correlation with isoprene in the model.**
- **Excluding ENSO in regions where it is highly correlated with VOCBB (Indonesia) or isoprene (India and southeast US).**
- **β_0: Trend term.**
- **$\beta_{1\ldots n}$: Regression coefficients (weights), indicating relative importance of each regressor.**

5.1. CH_2O IAV and trends in SON



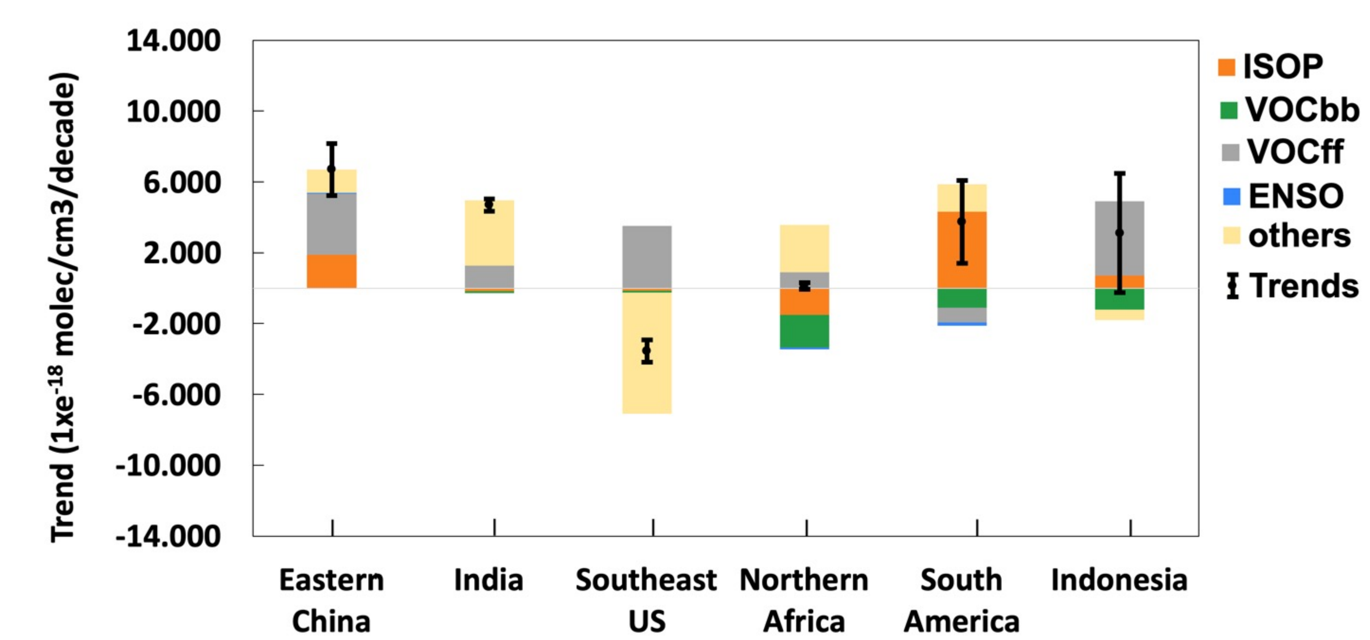
- **Model overestimates the observed CH_2O over Northern Africa and Southeast US, might having too much isoprene.**
- **The correlation between detrended time series (rIAV) increases in eastern China, northern Africa, South America.**

5.2. Source contributions to the IAV in CH_2O



- **Isoprene plays an important role in controlling the IAV of CH_2O.**
- **VOCBB is another crucial factor affecting IAV in three biomass burning regions.**
- **ENSO has a minor and negative impact on IAV over northern Africa and South America. It positively correlates to VOCBB over Indonesia, to isoprene over southeast US.**
- **Eastern China and India: changes in VOCFF and isoprene drive the CH_2O IAV.**

5.3. Source contributions to trends in CH_2O



- **VOCFF trends significantly impact CH_2O trend over Eastern China, Northern Africa and Indonesia.**
- **Isoprene leads to a positive trend in simulated CH_2O over South America, despite a negative VOCBB trend in the region.**
- **Sources other than isoprene, VOC emissions, and ENSO impact simulated trends in the Southeast US, India and Northern Africa.**

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