Increased propane emissions from the United States over the last decade

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Why Propane?

- It is the second most abundant non-methane hydrocarbon (NMHCs) after ethane
- It contributes to photochemical air pollution, including ozone and aerosol formation
- It is useful for distinguishing thermogenic from natural emissions of methane
- Recent atmospheric observations at remote stations suggest a reversal of earlier atmospheric declines [Helmig et al., 2016, Nature Geos.] that is largely due to increasing oil and natural gas production from the U.S. (Fig. 1)
- Reported production of propane has increased by a factor of 1.8 since 2011, primarily from Gulf Coast states (Fig. 2)

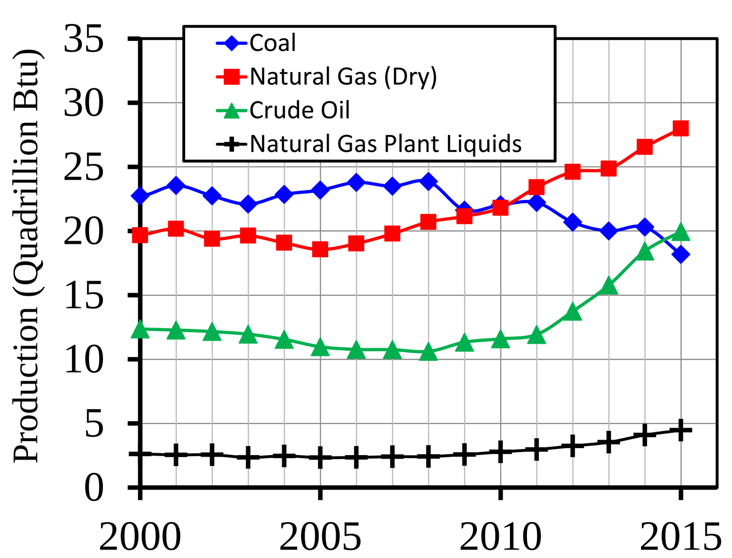


Fig. 1. Primary energy production from fossil fuels (US EIA)

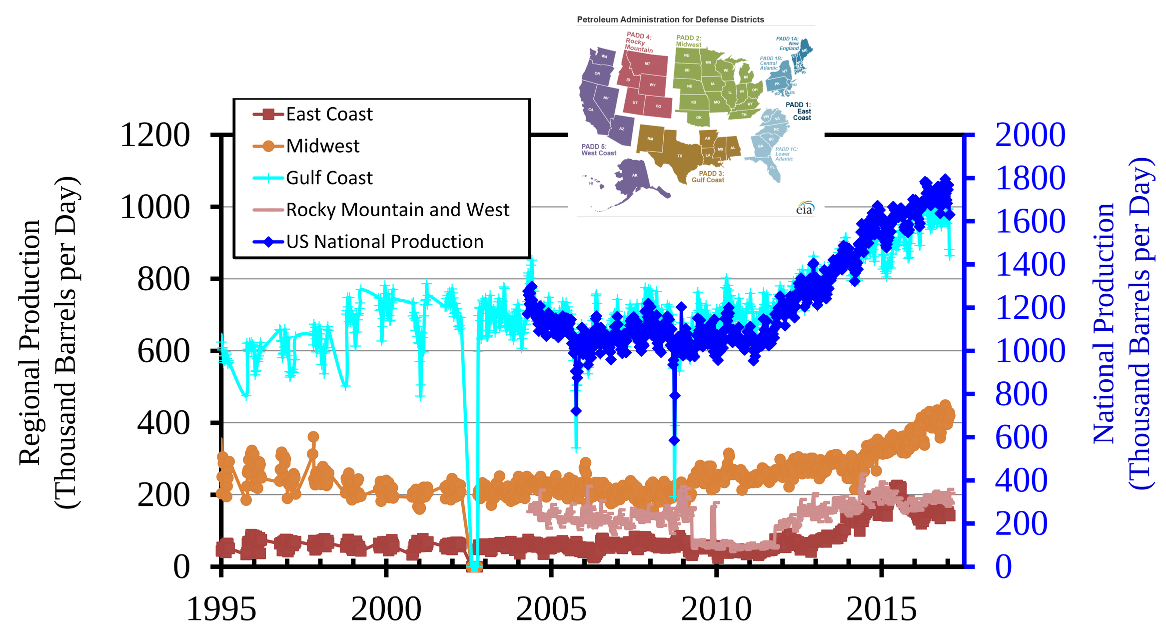


Fig. 2. Reported "net production" of propane and propene (US EIA)

Research Questions

- How much propane is emitted from the U.S. each year? What are the primary emitting sources of propane within the U.S.?
- Has propane emission increased as a result of increased oil and gas production? If so, by how much? From which region have emissions increased the most?

What have we already known about U.S. propane emissions?

From emissions inventories:

- U.S. emissions of propane are primarily emitted from anthropogenic sources (with estimated emissions of 0.1 – 0.7 Tg/yr). (Emissions from biomass burning and others were estimated at < 0.02 Tg/yr)
- C_3H_8 is mostly emitted from populated areas and from oil and gas production regions.
- Inventory-projected emissions after 2000 declined (Fig. 3).
- Higher emissions are expected during winter than summer (Fig. 3).

Previous regional atmosphere-based "top-down" studies suggest propane emissions:

- Smaller scale inventory estimates (State of Colorado) largely underestimate propane emissions in recent years (Petron et al., 2012, 2014)
- Propane emissions are primarily from natural gas production and processing, liquefied petroleum gas production, and geological seeps (Peischl *et al*., 2013; Wennberg *et al*., 2012)

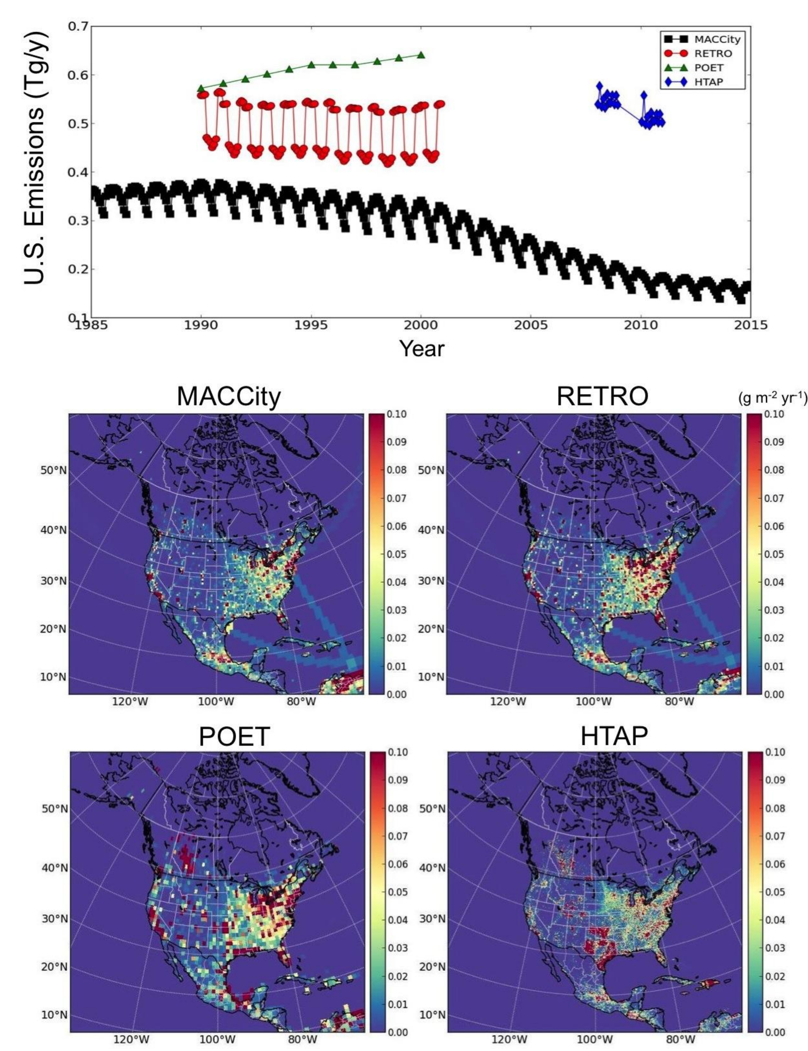


Fig. 3. U.S. anthropogenic propane emissions reported by inventories: MACCity, RETRO, POET, and HTAP.

Using inverse modeling of atmospheric data to infer propane emissions

- In general, this is how inverse models work for a chemically-inert tracer:

Atmospheric observations

$$\chi_{obs} = \chi_{bg} + Hs + \varepsilon \quad (1)$$

- χ_{obs}: Observed mole fractions
- χ_{bg}: Estimated background mole fractions
- H: Sensitivity of atmospheric observations to upstream surface fluxes, f(space, time)
- s: Upstream surface fluxes
- ε: Model-data mismatch errors

"First-guess" of surface fluxes

$$s = s_p + \varepsilon_p \quad (2)$$

- s: Upstream surface fluxes
- s_p: "first-guess" or prior surface fluxes
- ε_p: Errors of the "first-guess"

Posterior (or derived fluxes)

$$s = s_p + QH^T\left(HQH^T + R\right)^{-1}\left(z - Hs_p\right) \quad (3)$$

Here, $z = \chi_{obs} - \chi_{bg}$; R is the covariance matrix of ε; Q is the covariance matrix of ε_p

- **For propane, a chemically-active tracer:**

$$\chi_{obs} = \chi_{bg} - \chi_{chemloss} + \left(H_{corrected}\, s\right) + \varepsilon$$

$\left(H_{corrected}\, s\right)$: Enhancements due to upstream surface emissions, but corrected for chemical loss that occurs during transport to a sampling location (achieved through a modification of the surface sensitivity "H")

Rewritten as:

$$\chi_{obs} = \chi_{bg} \exp\left(\int_0^{t_1} -\kappa[OH]dt\right) + H \exp\left(\int_0^{t_2} -\kappa[OH]dt\right) s + \varepsilon$$

Using this equation substituting Eq. (1) and keeping the rest of equations the same yield the posterior solution of propane emissions with OH loss included

Observational Evidence for Increased Propane Emissions from the U.S.

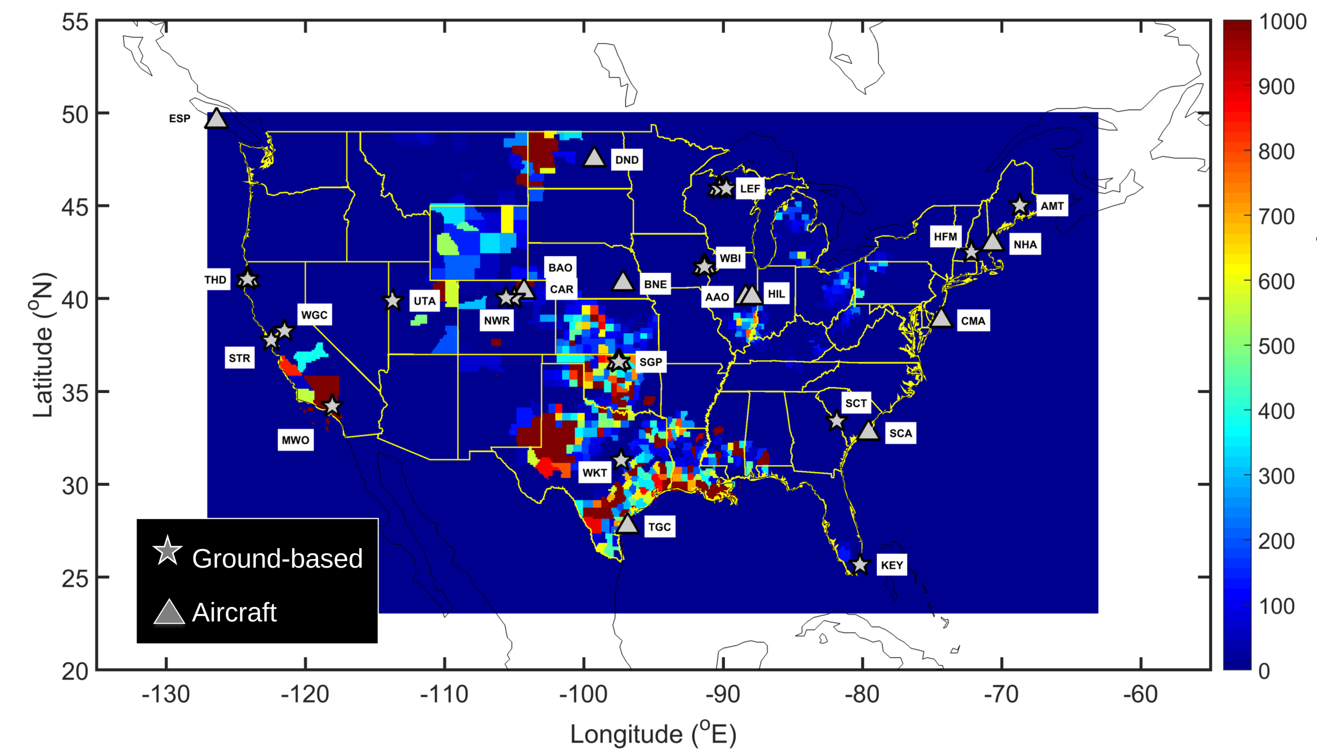


Fig. 4. NOAA's cooperative air sampling network over North America. Color shading indicates oil production magnitude in the U.S. for 2011 (U.S.D.A.)

- Larger mole fractions of C_3H_8 observed at sites in the CONUS relative to those at remote sites, confirming C_3H_8 emissions from the U.S.
- Large positive trends were observed at sites strongly influenced by oil and gas production, indicating likely increased C_3H_8 emissions from oil and gas production activities

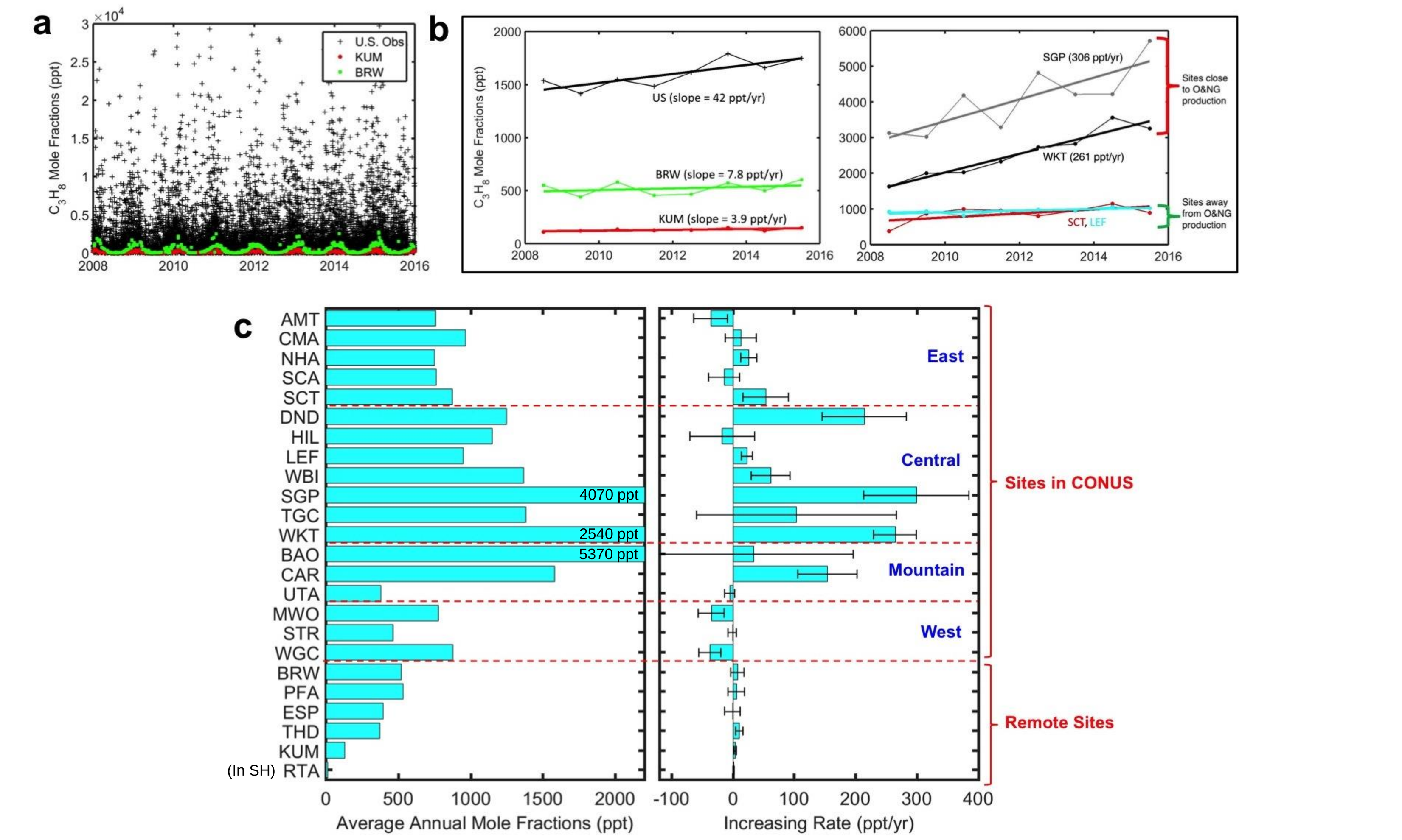


Fig. 5. (a) C_3H_8 mole fractions observed in the CONUS and in remote atmosphere. (b) Annual mole fraction trends observed in the CONUS and remote atmosphere. (c) Average annual mole fractions and trends in annual mole fractions of C_3H_8 observed at sites in the CONUS and remote atmosphere.

Propane Emissions and Emission Trend Inferred from Atmospheric Observations

Spatial Distribution:

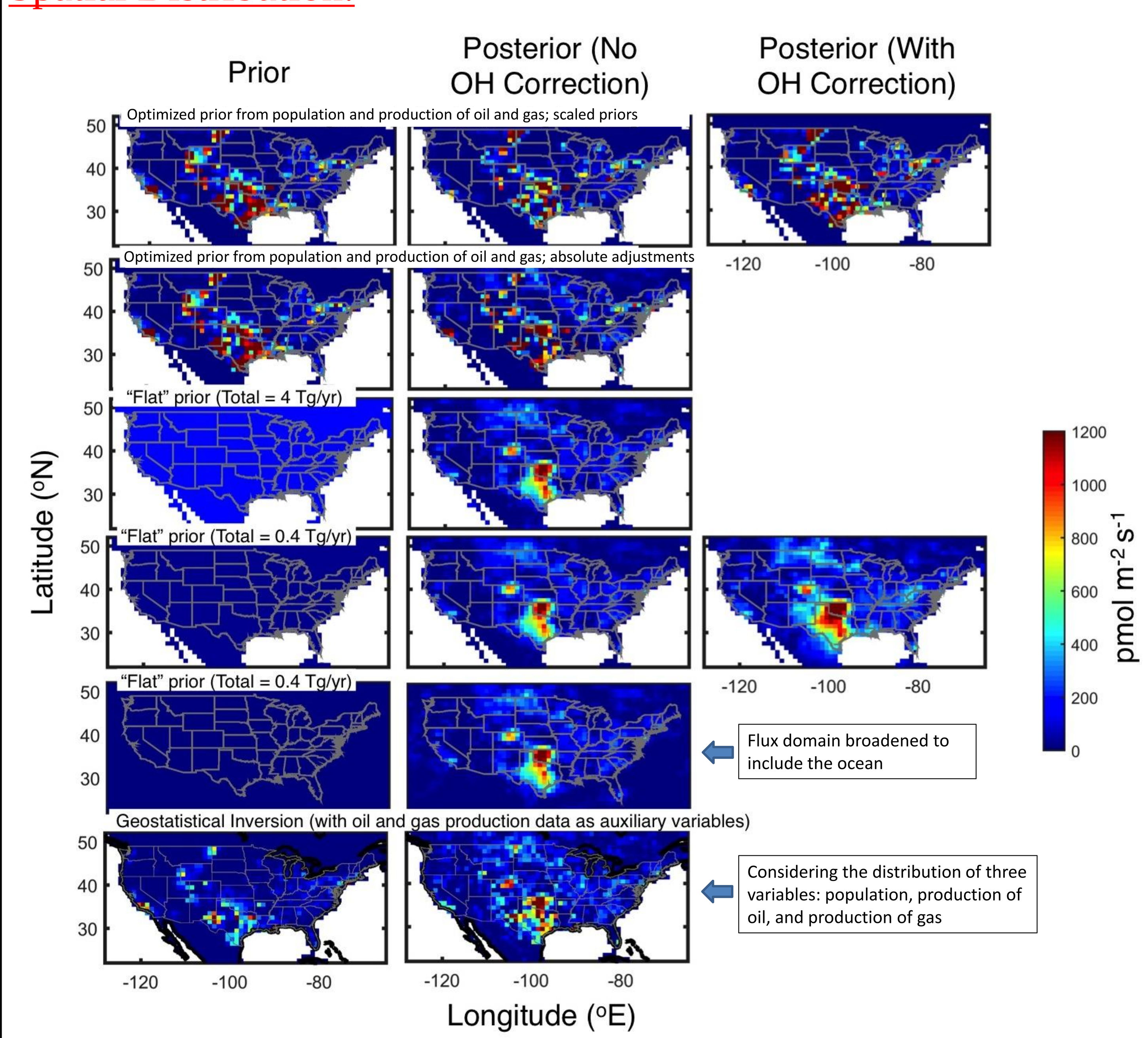


Fig. 6. U.S. emissions of propane derived from Bayesian Inversions with different priors and a Geostatistical Inversion (GI).

- Results indicate the majority of C_3H_8 emissions are from oil and gas production regions; but emissions from populated areas are also important to explain the atmospheric observations as shown in the Bayesian Information Criterion (BIC) test.

Spatial Models	BIC Scores	ΔBIC (relative to the lowest BIC score)
oil	74466	162
gas	74533	229
population	74732	428
oil+gas	74489	185
gas+population	74489	185
oil+gas+population	74304	0

Note that, lower BIC score indicates better spatial model

Seasonality:

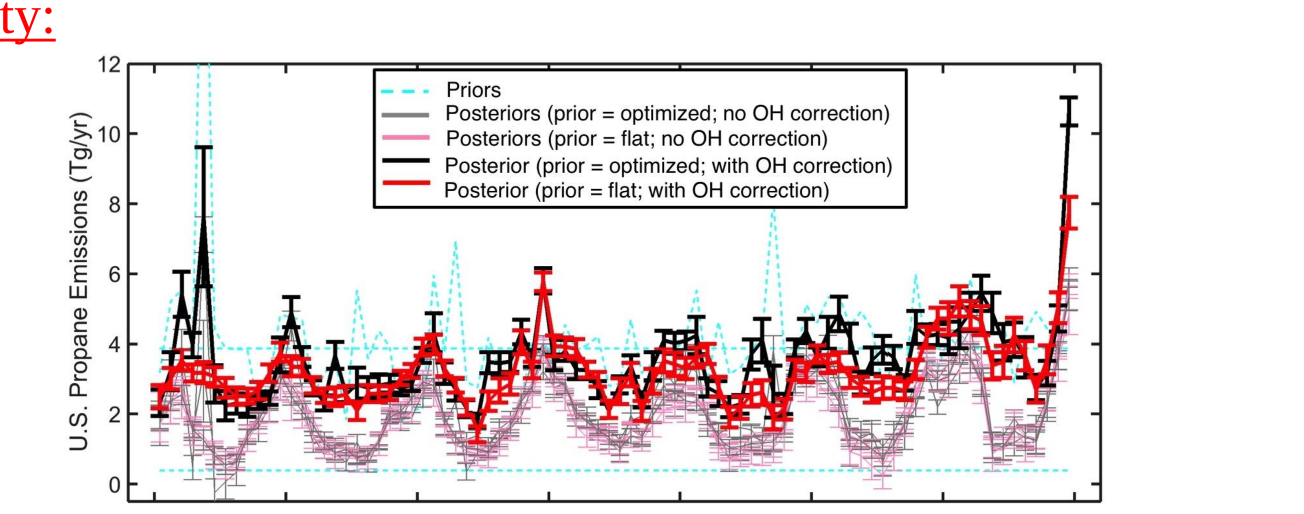
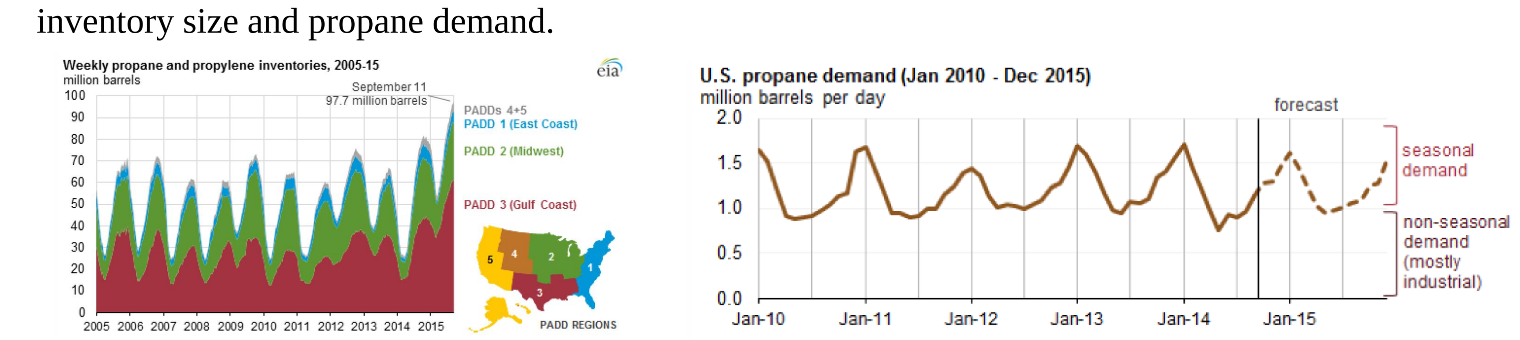
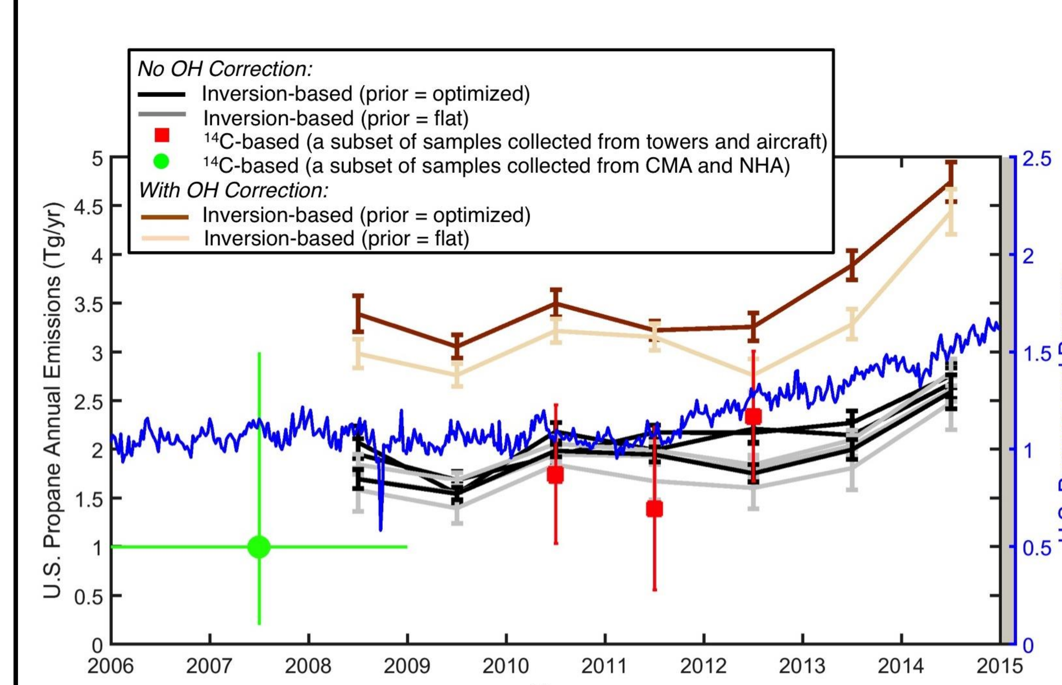


Fig. 7. Derived monthly propane emissions from different priors and inverse modeling methods. ***Higher winter emissions than summer emissions are derived***, with seasonal variations of propane/propene inventory size and propane demand.



Annual Emissions and Emission Trend:



* Estimated propane emissions show an increase from 2011 to 2014, coincident with the increase of propane/propene production (blue line) in the U.S. This U.S.-related emission increase contributes to the observed recent increases in propane atmospheric concentrations at remote stations (Helmig *et al*., 2016).
* Considering OH losses of propane substantially increases derived emission magnitudes (by a factor of 2).
* Observationally-derived national propane emissions are about 10 times higher than those projected by the MACCity inventory for the same time period.

Fig. 8. U.S. annual emissions of propane derived from inverse analyses of observations for 2008 – 2014 with and without considering OH chemistry. Propane emissions estimated from a ^{14}C-based tracer ratio method for 2010 – 2012 from a subset of air samples collected from towers and aircraft in a previous study only using data from two east coastal sites (Miller et al., 2012) are also shown.