Interpreting satellite NO_2 observations over the US: the importance of accounting for the free tropospheric background

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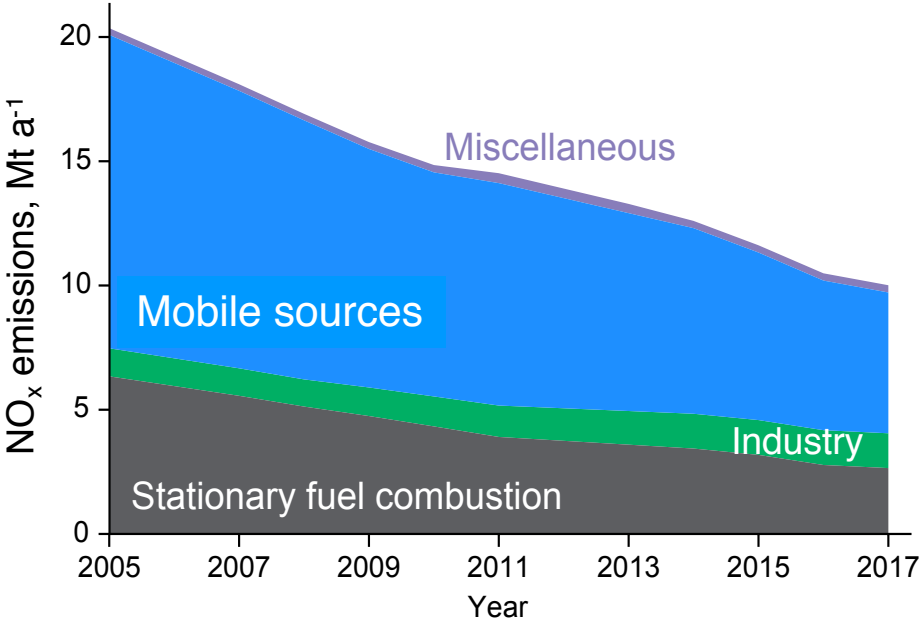
Silvern, R.F., D.J. Jacob, L.J. Mickley, M.P. Sulprizio, K.R. Travis, E.A. Marais, R.C. Cohen, J.L. Laughner, S. Choi, J. Joiner, and L.N. Lamsal, Using satellite observations of tropospheric NO2 columns to infer long-term trends in US NOx emissions: the importance of accounting for the free tropospheric NO2 background, Atmos. Chem. Phys., 19, 8863–8878, 2019.

US NO_x emissions have continued to decrease according to EPA… but OMI tropospheric NO_2 column observations suggest otherwise!

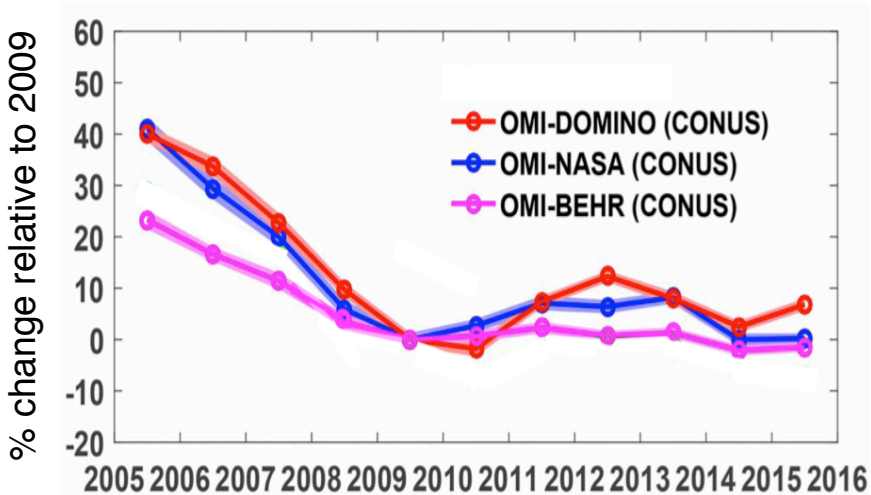
EPA National Emission Inventory (NEI): 53% sustained decrease of NO_x emissions over 2005-2017

EPA, 2018

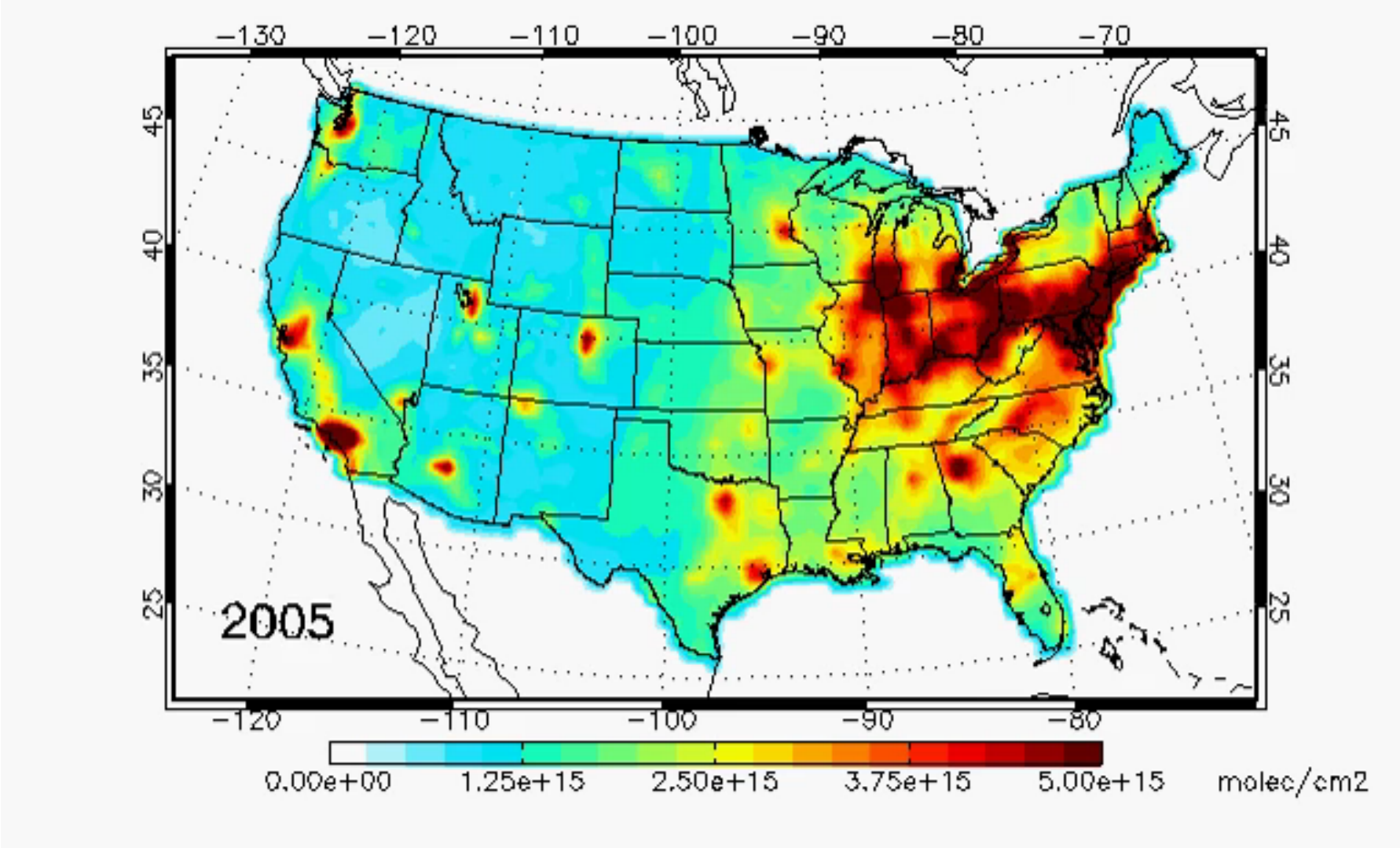


OMI NO_2 columns over CONUS, 2005-2015: flat after 2009

Jiang et al., PNAS 2018



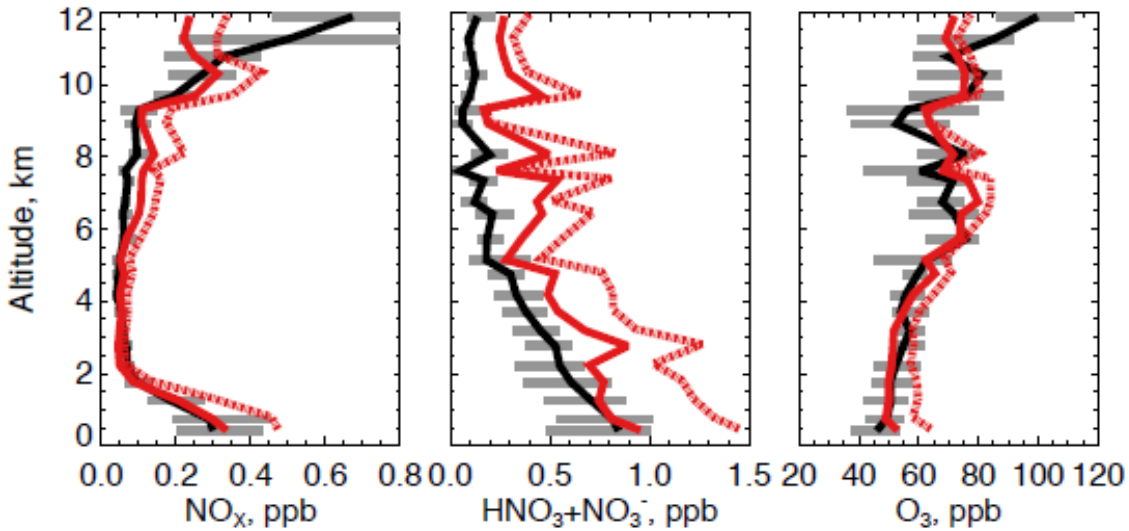
OMI annual NO_2 trends over CONUS, 2005-2017



Hypothesis #1:
EPA NO_x emission trends are wrong

Mobile sources in EPA inventory are too high, so maybe trends are wrong as well?

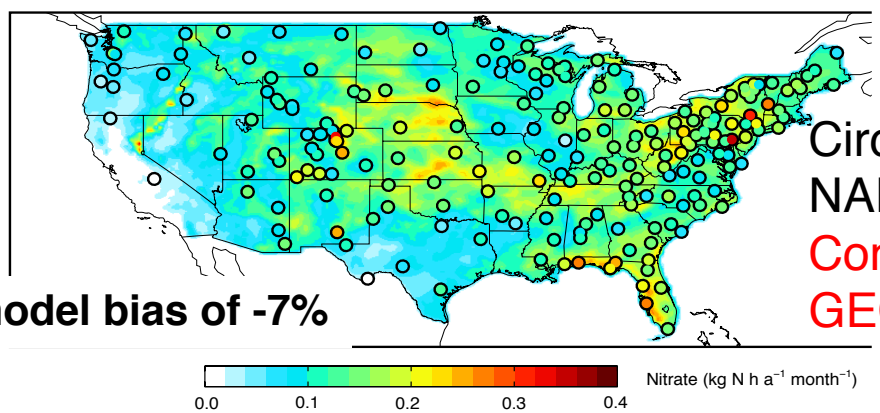
NASA SEAC^{4}RS aircraft campaign, Aug Sep 2013



Observed
GEOS-Chem
with EPA NO_x emissions
with EPA NO_x emissions/2

Travis et al., ACP 2016

nitrate wet deposition fluxes, Aug-Sep 2013

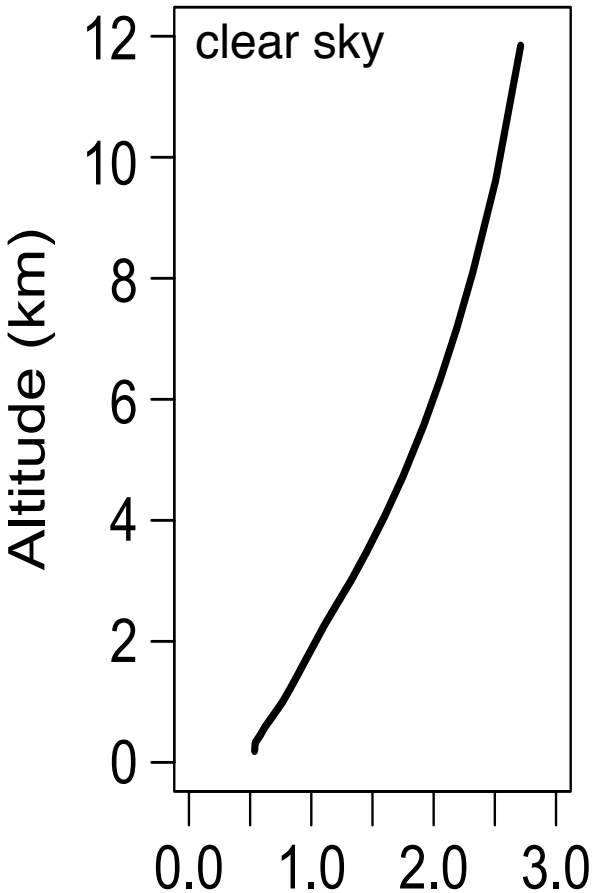


model bias of -7%

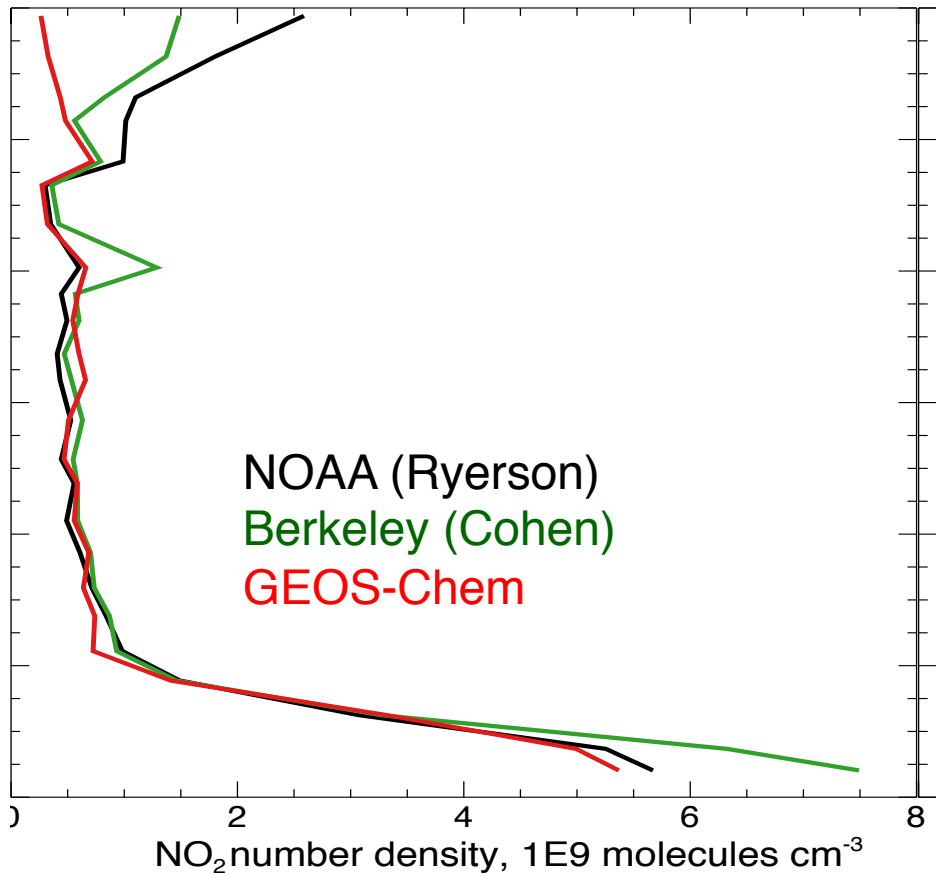
Circles:
NADP observations
Contours:
GEOS-Chem with EPA emissions/2

Hypothesis #2:
US NO_x emissions are now so low that OMI mostly sees background

OMI NO_2 sensitivity vs. altitude



Mean SEAC^{4}RS NO_2 profile over Southeast

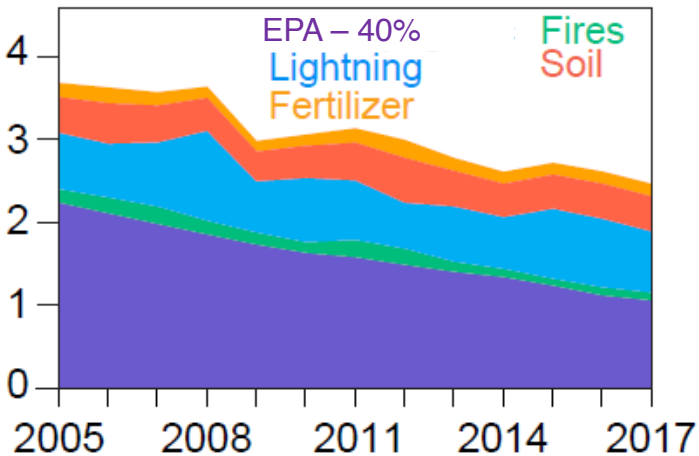


Boundary layer below 2 km accounts for only 20-35% of tropospheric NO_2 column as seen by OMI

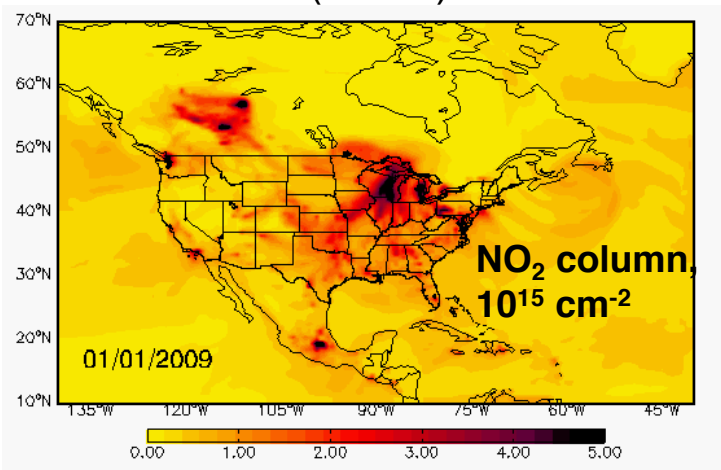
Travis et al. , ACP 2016; Silvern et al., GRL 2018

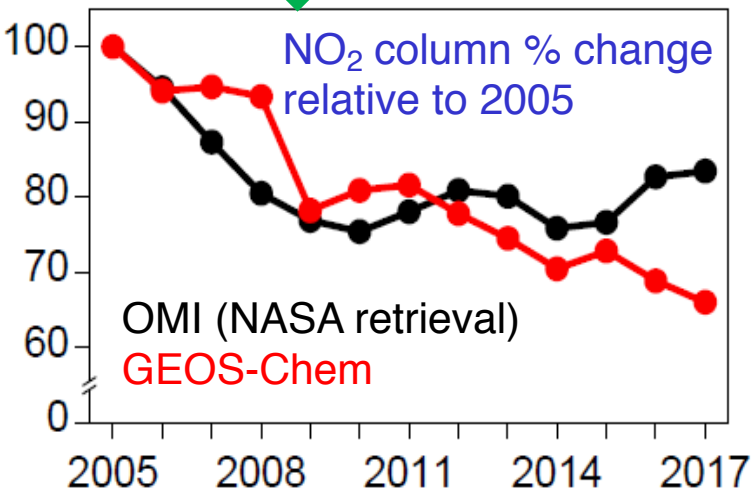
Use GEOS-Chem to interpret trends from different data sets

GEOS-Chem NO_x emissions
(Tg N a^{-1})



2005-2017 GEOS-Chem simulation
0.5°x0.625° (50 km) resolution

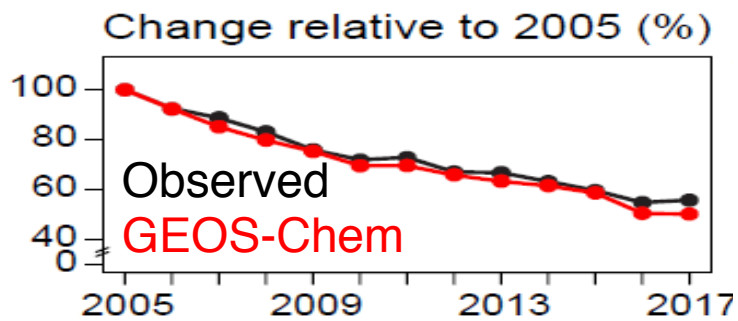




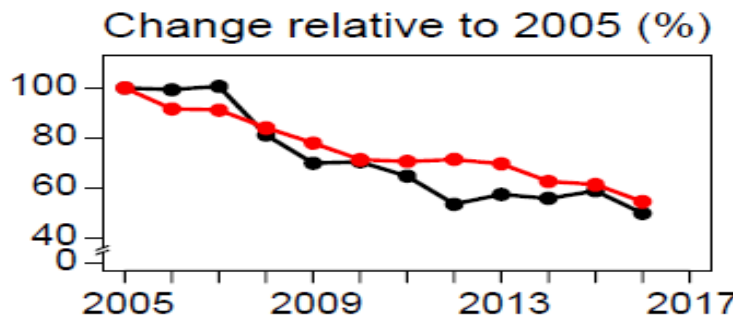
- GEOS-Chem driven by EPA emission trend does not reproduce flat OMI trend after 2009
- GEOS-Chem NO_x tropospheric column lifetime decreases by 5% from 2005 to 2017

2005-2017 trends in surface data

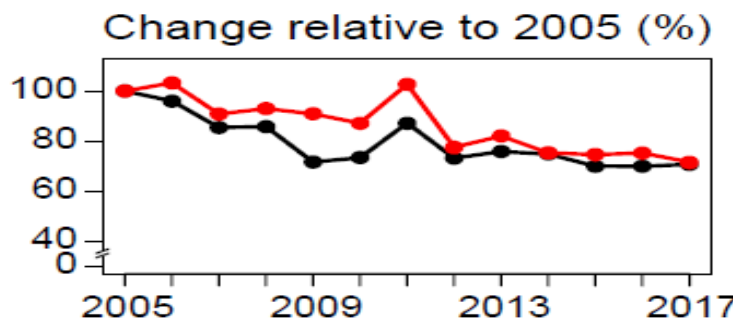
AQS NO_2 (urban)



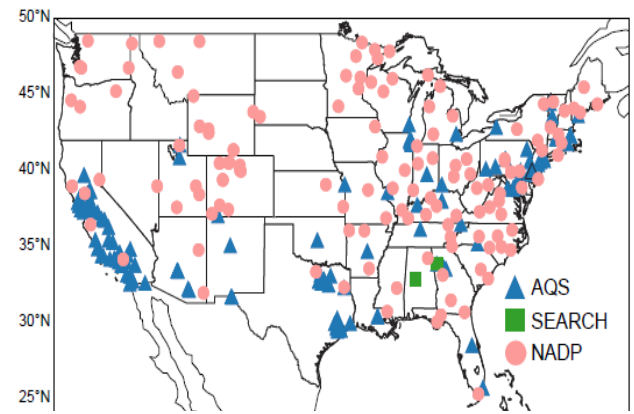
SEARCH NO_2 (2 rural sites)



NADP nitrate wet deposition



Surface sites with continuous annual records for 2005-2017

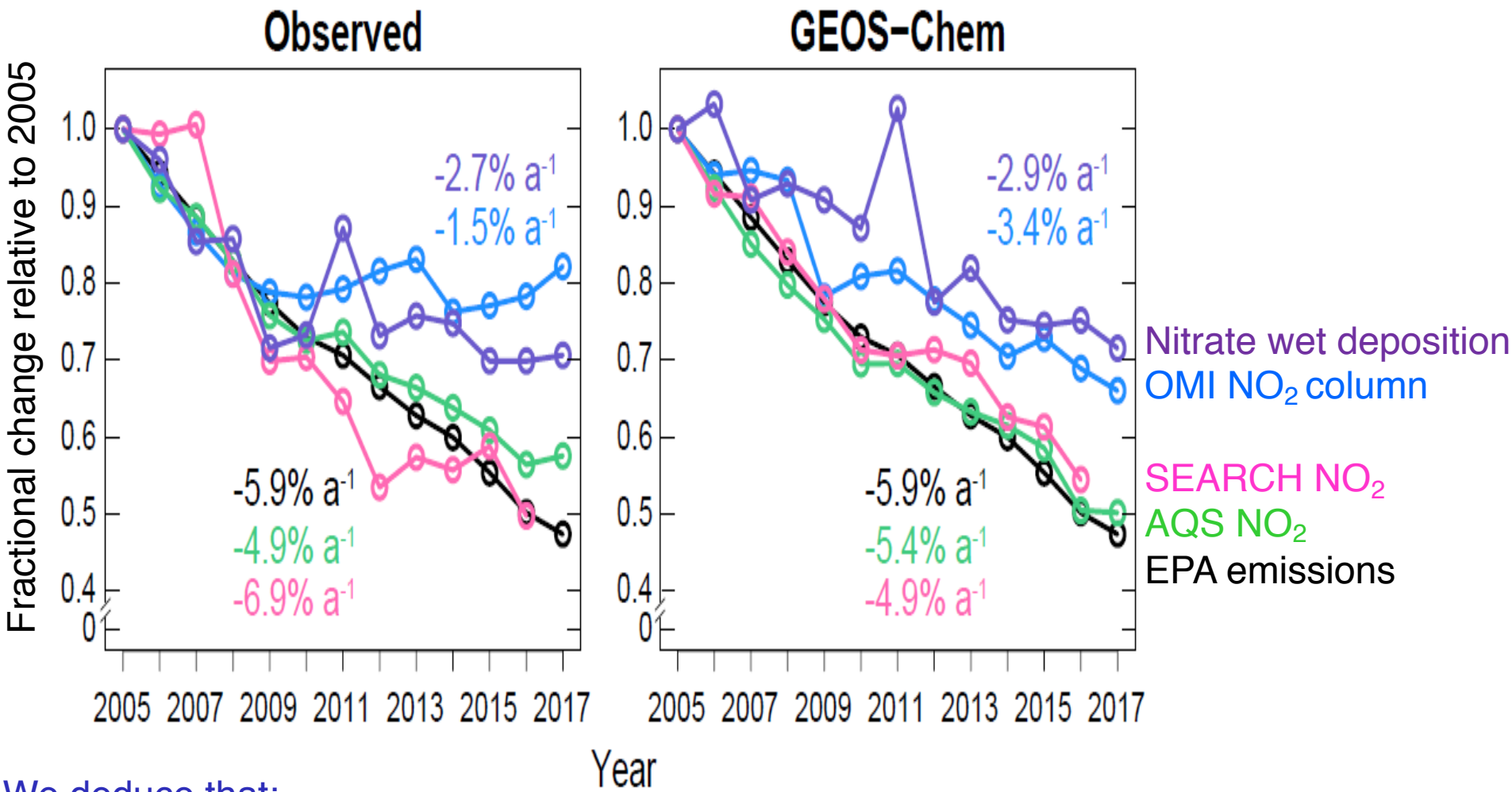


GEOS-Chem matches the observed relative trends:

- surface NO_2 is most sensitive to anthropogenic emissions
- nitrate deposition is sensitive to background (soils, lightning, transport from outside US)

US anthropogenic sources contribute only 50% of nitrate wet deposition in 2005, 31% in 2017

Comparing 2005-2017 relative trends for the different quantities

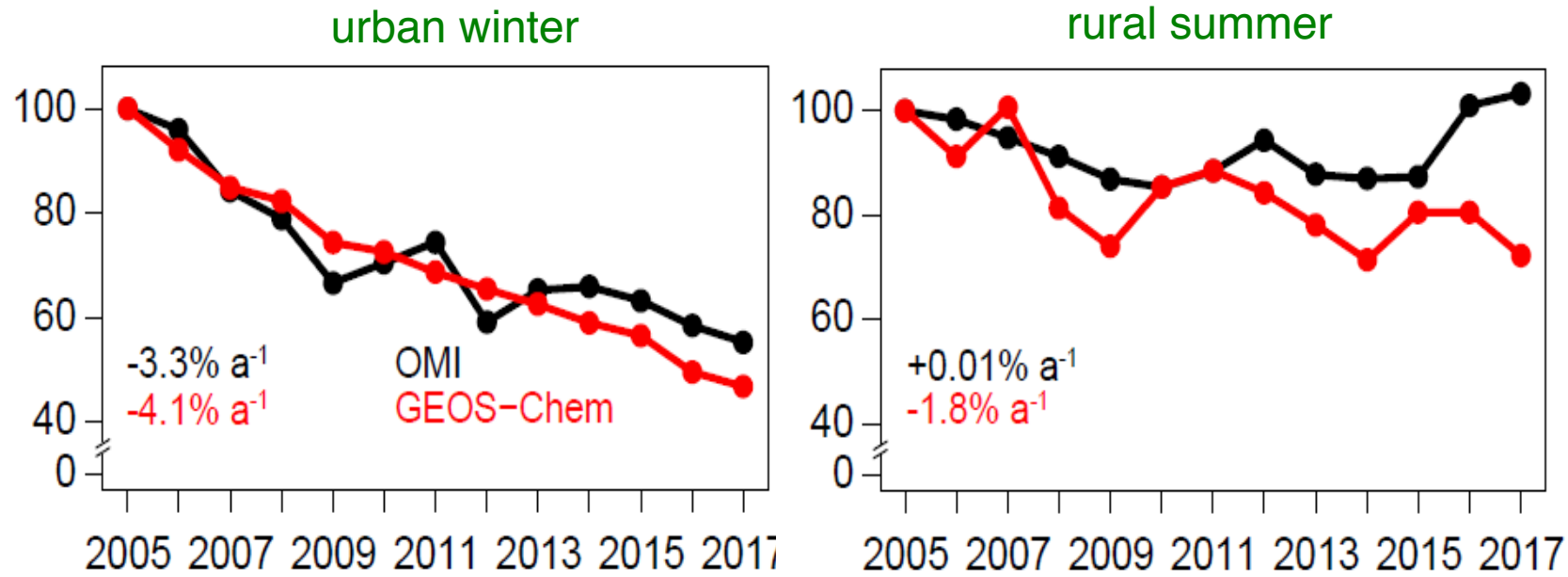


We deduce that:

- EPA 2005-2017 trend in NO_x emissions is largely correct
- OMI NO_2 columns have strong background influence
- GEOS-Chem underestimates free tropospheric background NO_2

Separating OMI trends by urban vs. rural, winter vs. summer

Trends in NO_2 columns relative to 2005 (%)



OMI shows:

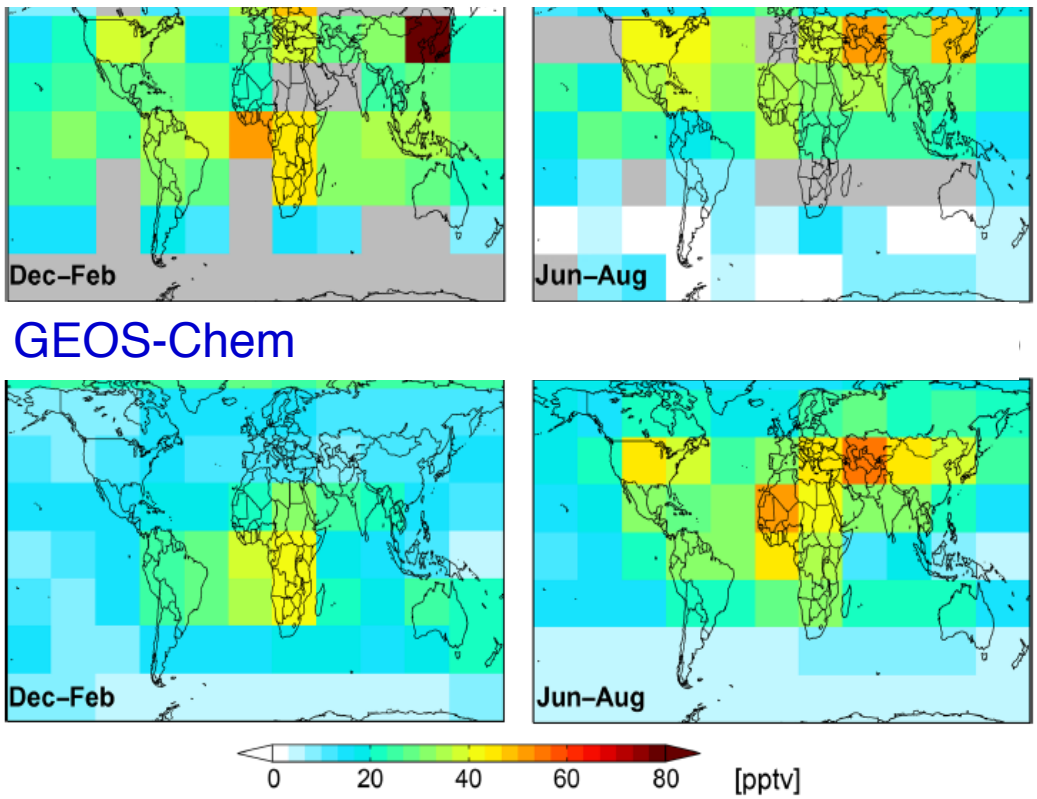
- steady decrease in urban winter where background is relatively low
- no trend in rural summer where background is relatively high

OMI NO_2 is useful to detect hotspots and plumes,
but inference of regional emissions is far more uncertain

Silvern et al., ACPD 2019

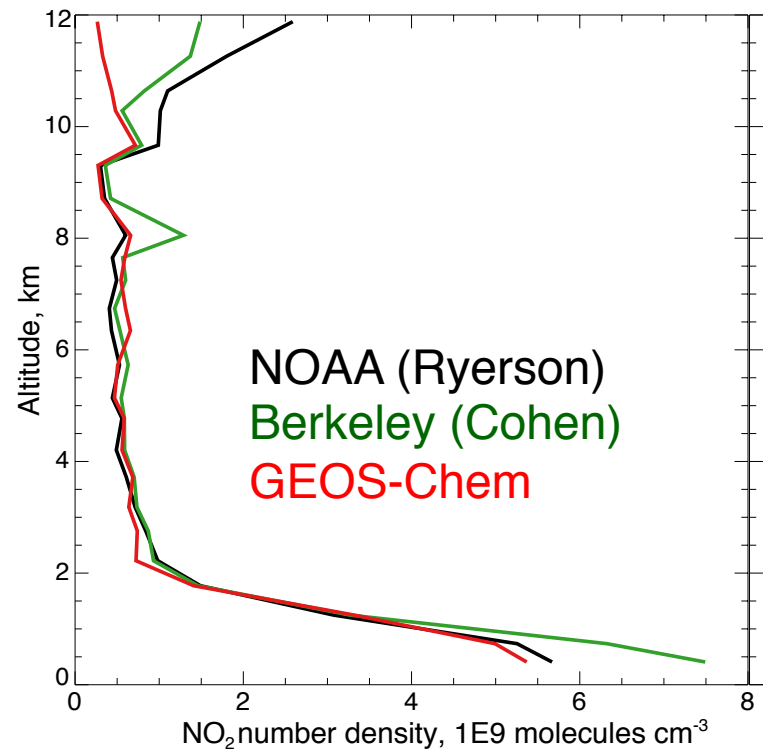
GEOS-Chem underestimates free tropospheric NO_2 in winter, upper tropospheric NO_2 in summer

OMI cloud-sliced free tropospheric NO_2 (6-10 km):
Sungyeon Choi and Joanna Joiner, NASA



Marais et al. [ACP 2018]

NO_2 vertical profiles in SEAC^{4}RS (Aug-Sep 2013)



Could high NO_2 observed in upper troposphere extend into the lowermost stratosphere?

Conclusions

- Anthropogenic NO_x emissions in the US have decreased steadily over the 2005-2017 period, even though mean OMI NO_2 values over the US show no trend after 2009

- US NO_x emissions are now so low that satellite measurements of tropospheric NO_2 columns over the US are dominated by background sources (other than US anthropogenic) except in urban areas

- Better understanding of the free tropospheric NO_2 background is needed to interpret satellite NO_2 tropospheric column data in terms of NO_x emissions and their trends