Magnitude and trend of NO$_x$ and SO$_2$ emissions constrained by OMI observations

Zhen Qu and Daven K. Henze
University of Colorado Boulder

zhen.qu@colorado.edu
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Two ways to estimate emissions

• Bottom-up estimates – prior emissions:
  - Emission = emission factor x activity
  - Large uncertainties, lag in time

• Top-down estimates – posterior emissions
  - Use observations and physical model to solve inverse problem which gives the maximum likelihood estimate of emissions
Top-down emission studies

- Analytic inversion
  - Expensive to compute the Jacobian matrix;
  - Approximated by linear relationships of NO$_2$ column to NO$_x$.

(Konovalov et al., 2006, 2008)
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• Ensemble Kalman Filter
  - Updated error covariance matrix;
  - Expensive using large ensemble members;
  - Hard to implement realistic localization.

  (Miyazaki et al., 2015, 2017)

US NO$_x$ emissions

A posteriori
EDGAR-HTAP v2
A priori
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• **Plume model**
  - Identify large, isolated, point sources;
  - Need average over multiple years.

  *(Beirle et al., 2011)*
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• **Mass balance**
  - Fast;
  - Approximate transport & nonlinear chemistry.

  \[(\text{Martin et al., 2003})\]
History of 4D-Var NO$_x$ emission estimates

1999

Elbern & Schmidt
Full 4D-Var for 3D CTM
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Elbern et al.
Pseudo observation

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NO emission rates can be estimated using O$_3$ observations
History of 4D-Var NO\textsubscript{x} emission estimates

Elbern et al.
Pseudo observation

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Muller & Stavrakou
GOME NO\textsubscript{2}

2005

First use of NO\textsubscript{2} satellite observations
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- **2000**
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- **2005**
  - Elbern et al.: Pseudo observation

- **2009**
  - Henze et al.: IMPROVE aerosol

First use of surface aerosol measurements
History of 4D-Var NO\textsubscript{x} emission estimates

1999

Elbern & Schmidt
Pseudo observation

2000

Full 4D-Var for 3D CTM

2005

Elbern et al.

2009

Henze et al.
IMPROVE aerosol

2013

Xu et al.
MODIS radiance

\text{NO}_x\text{ emission (posterior – prior)/prior}
Elbern & Schmidt
Full 4D-Var for 3D CTM

Henze et al.
IMPROVE aerosol

Qu et al.
OMI NO₂

Elbern et al.
Pseudo observation

Muller & Stavrakou
GOME NO₂

History of 4D-Var NOₓ emission estimates

1999

2000

2005

2009

2013

2017

NOₓ emission scaling factors
• **Model:** GEOS-Chem chemical transport model and its adjoint
  - Meteorological input from Goddard Earth Observing System (GEOS)
  - Prior emissions: HTAP v2.1 bottom-up inventory (2010) for all years and domains
  - Global domain: 2° lat x 2.5° lon resolution for 2005 – 2017

**Surface NO\textsubscript{x} concentration (Jan 2010)**

- Global 2° x 2.5°
  - 2005 – 2017
  - MERRA - 2
Model setups: 3 domains

Surface NO$_x$ concentration (Jan 2010)

- Nested US and nested EA domain: BC from global 4° x 5° simulation, 2005 - 2012
Satellite observation

- Ozone Monitoring Instrument (OMI) onboard Aura: NO$_2$ and SO$_2$
- Overpass time: 13:45 local time, daily global coverage
- Footprint: 13 km x 24 km

- Use level 2 product for all work in the presentation
- Column density: total NO$_2$ and SO$_2$ molecules from surface to the top of the atmosphere within a model grid [molec cm$^{-2}$]
Methods

- **Inversion approaches:**

  4D-Var:
  - adjust emissions independently in each grid cell
  - takes into account transport and chemical reactions
  - computationally expensive
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Hybrid 4D-Var / Mass balance:
- blend of accuracy and efficiency
Outline

1. Top-down NO\textsubscript{x} emissions
2. Top-down SO\textsubscript{2} emissions
3. Joint NO\textsubscript{x} and SO\textsubscript{2} inversions
4. Sector-based inversion
Hybrid inversion for NO\textsubscript{x}

Hybrid method:
Base year (2010): 4D-Var

Scaled emissions in pseudo observation test

- Hybrid posterior has smaller NMSE (by 59\% to 78\%) and better correlation.

(Qu et al., 2017)
Differences between bottom-up and top-down estimates

Top-down – bottom-up, 2010 [TgN/year]

- Underestimates in HTAP at regions with large anthropogenic sources (East Coast of US & Mexico City)
- Overestimates in HTAP at regions with moderate anthropogenic sources (mid US)
Smaller seasonality of top-down NO\textsubscript{x} emissions

US NO\textsubscript{x} emissions in 2010

![Graph showing US NO\textsubscript{x} emissions in 2010 with two curves: Prior (black) and Posterior (red).]
Inter-annual variation: Changes of NO\textsubscript{x} emissions in NA

2012 – 2005 annual NO\textsubscript{x} budget [TgN]

- Annual budget of top-down NO\textsubscript{x} emissions decrease by 20% from 2005 to 2012 in the US
- NO\textsubscript{x} emission changes in Mexico are less than 1% from 2005 to 2012
Large differences in OMI NO$_2$ column from two retrievals
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OMI VCD$_{SP}$ – VCD$_{DOMINO}$, Jan 2015

Vertical Column Density:
Standard Product (SP): VCD$_{SP} = $ VCD$_{SP_OMI} \times$ AMF$_{SP}$/AMF$_{GC_SP}$
DOMINO Product: VCD$_{DOMINO} = $ VCD$_{DOMINO_OMI} \times$ AMF$_{DOMINO}$/AMF$_{GC_DOMINO}$

- NO$_2$ column densities from SP are ~ 50% smaller than that from DOMINO in densely populated and industrial regions. (Qu et al., 2017; Canty et al., 2015; Zheng et al., 2014)
Different magnitude of NO\textsubscript{x} emissions from NASA SP and DOMINO retrievals

- Posterior NO\textsubscript{x} emissions from SP is smaller than that from DOMINO by 39-46%.
Different magnitude of NO\textsubscript{x} emissions from NASA SP and DOMINO retrievals

Total NO\textsubscript{x} emissions in the US

- Posterior NO\textsubscript{x} emissions from SP is smaller than that from DOMINO by 39-50%.
- The slowdown of NO\textsubscript{x} emissions is not reflected in NEI inventory.

(Jiang et al., 2018)
Different magnitude of NO$_x$ emissions from NASA SP and DOMINO retrievals

Total NO$_x$ emissions in Mexico

Figure 4-7. Annual Average Satellite NO$_2$ Columns over Mexico.

(RAMBOLL report)
Different magnitude of NO\textsubscript{x} emissions from NASA SP and DOMINO retrievals

Total NO\textsubscript{x} emissions in Mexico

![Annual Average Satellite NO\textsubscript{2} Average Over Mexico](chart1)

![NO\textsubscript{x} emissions](chart2)

- Posterior NO\textsubscript{x} emissions from SP is smaller than that from DOMINO by 47-51%.

(RAMBOLL report)
Impact of assimilating NO$_2$ observations on O$_3$ (2010)

Surface O$_3$ concentration (posterior NO$_x$ – prior NO$_x$) [ppbv]

- NO$_x$ emission is overestimated in US bottom-up inventory
- Simulated O$_3$ are generally overestimated in US using HTAP 2010 emissions
Impact of assimilation on improving estimates of surface $O_3$ depends upon the $O_3$ metric, emphasizing the importance of hourly $NO_x$ constraints.

NMB of summertime surface $O_3$ (2010, compared to TOAR)

- Posterior simulations have smaller NMB and NMSE and better seasonality and inter-annual variation in 24 hour $O_3$. 
SO$_2$ emissions constrained by OMI SO$_2$ NASA and BIRA products

- **3 OMI SO$_2$ products**: NASA standard (SP), NASA prototype, BIRA

  Treatment of clouds, radiative transfer model, and retrieval algorithm lead to differences in NASA and BIRA SO$_2$ retrievals, which are more consistent when VZA and SZA are small

(Qu et al., 2019a)
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- **SO$_2$ emissions continuously increase in India from 2005 – 2017 and start to decrease in China from 2008.**

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• **SO$_2$ emissions continuously increase in India from 2005 – 2017 and start to decrease in China from 2008.**

• **Evaluation with surface & aircraft measurements**: Reduced NMB in annual mean surface SO$_2$ in China, India and US but not in Korea possibly due to differences in SO$_2$ vertical profile in model and real atmosphere.  
  
  *(Qu et al., 2019a)*
Top-down emissions

Still ...

- Chemical interactions are not being considered so far
- Uncertainties in other species emissions are likely degrading the top-down emission of the constrained species
Joint NO$_2$ & SO$_2$ 4D-Var inversion -- better match observations and surface measurements (January, 2010)

Joint – Single posterior emissions

Joint: assimilate NO$_2$ and SO$_2$ observations to optimize NO$_x$ and SO$_2$ emissions simultaneously

Single: only assimilate NO$_2$ (SO$_2$) observations to optimize NO$_x$ (SO$_2$) emissions

(Qu et al., 2019b)
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SO$_2$ columns (GEOS-Chem – OMI)

\( [\text{DU}] \)

\(-1 \quad -0.33 \quad 0.33 \quad 1\)

\( [10^6 \text{ kgS box}^{-1} \text{ mon}^{-1}] \)

\( [-1 \quad -0.33 \quad 0.33 \quad 1] \)

\( [10^{11} \text{ molec cm}^{-2} \text{ s}^{-1}] \)

(Qu et al., 2019b)
Similar magnitude and trend of single species and joint inversion posterior emissions

China $SO_2$ emissions [TgS]  
China $NO_x$ emissions [TgN]

(Qu et al., 2019b)
Accounting for correlated co-emitted pollutants in 4D-Var

Transportation

Energy

Sector-based emission scaling factor

Similar ratio of NO\textsubscript{x}, SO\textsubscript{2} and CO emissions in the same sector, yet very different across sectors. (Qu et al., in prep)

Assimilate:

MOPITT CO
OMI NO\textsubscript{2}
OMI SO\textsubscript{2}
Evaluations of posterior simulations with measurements

- NMB of posterior simulations from sector-based inversions are 59.8% ($\text{SO}_2$) and 61.4% ($\text{NO}_2$) smaller than the ones from species-based inversion.
Summary

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• Reduced error in NO$_x$ and SO$_2$ top-down emissions using multiple species joint inversion, through correction of OH concentration in the model, at months when observation uncertainties of optimized species are large.
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• Reduced error in NO\textsubscript{x} and SO\textsubscript{2} top-down emissions using multiple species joint inversion, through correction of OH concentration in the model, at months when observation uncertainties of optimized species are large.
• A new sector-based inversion is developed to estimate emissions at process level using satellite observations.

Qu et al. (2019a), SO\textsubscript{2} emission estimates using OMI SO\textsubscript{2} retrievals for 2005 – 2017
Qu et al. (2019b), Hybrid mass balance / 4D-Var joint inversion of NO\textsubscript{x} and SO\textsubscript{2} emissions in East Asia
Qu et al. (2017), Monthly top-down Nox emissions for China (2005-2012): A hybrid inversion method and trend analysis
Causes of slowdown

• The decreasing relative contributions of gasoline cars, due to the ongoing effectiveness of three-way catalytic converters

• The increasing relative emissions of NO$_x$ from off-road vehicles and industrial, residential, and commercial boilers

• Slower-than expected reductions in emissions by heavy-duty diesel trucks that have newer (and still maturing) catalytic converter technologies

(Jiang et al., PNAS, 2018)