Deriving top-down NO$_x$ and SO$_2$ emissions simultaneously using OMI observations and GEOS-Chem adjoint

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Satellite observations provide timely update of pollutant emissions and concentrations

\( \text{NO}_2 \) column density for US

(Jiang et al., 2018; Qu et al., 2017; Miyazaki et al., 2017; Ding et al., 2017; Krotkov et al., 2016; de Foy et al., 2016; Liu et al., 2016; Duncan et al., 2016; Cui et al., 2016)

- OMI shows unexpected slow down of \( \text{NO}_2 \) concentration in US
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(NO₂ column density for US) (Top-down SO₂ emissions for China)

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- SO₂ emissions starts to decrease from 2007 in China.

(Koukouli et al., 2017; Li et al., 2017; Krotkov et al., 2016; Wang et al., 2016)
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**NO₂ column density for US**

**Top-down SO₂ emissions for China**

(Jiang et al., 2018; Qu et al., 2017; Miyazaki et al., 2017; Ding et al., 2017; Krotkov et al., 2016; de Foy et al., 2016; Liu et al., 2016; Duncan et al., 2016; Cui et al., 2016)

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- OMI shows unexpected slow down of NO₂ concentration in US
- SO₂ emissions starts to decrease from 2007 in China.
- Uncertainties: overlooked chemical interactions & retrieval method
Improved performance in emission estimates using multi-species observation and optimization

- Better performance of joint 4D-Var (by 8.7%) and mass balance (by 5.1%) than single species inversion

**Pseudo observation test**

**Legend**
- **VarJ**: 4D-Var jointly
- **VarS**: Sum of two individual 4D-Var
- **MBJ**: Mass balance jointly
- **MBS**: Sum of two individual mass balance
- **OptS**: Optimize SO$_2$ emissions
- **OptN**: Optimize NO$_x$ emissions
- **ObsS**: Observe SO$_2$ column
- **ObsN**: Observe NO$_2$ column
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Improved performance in emission estimates using multi-species observation and optimization

- Better performance of joint 4D-Var (by 8.7%) and mass balance (by 5.1%) than single species inversion
- Largest decrease of NMSE if observe and optimize both species at the same time
Generally reduced error and bias in hybrid joint posterior NO$_x$

Setup (2005-2012):

- **True emissions**: 10% annual growth rate
- **Prior emissions**: 2010 true emissions x random noises x 0.5, for all years
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- Hybrid approach reduce error (bias) by 17-53% (17-83%) compared to MB for $\text{NO}_x$, except for 2006
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- Hybrid approach reduce error (bias) by 17-53% (17-83%) compared to MB for NO\textsubscript{x}, except for 2006
- Joint inversion reduce error (bias) by 0-12% (3-13%) compared to single species inversion for NO\textsubscript{x}
Generally reduced error and bias in hybrid joint posterior SO$_2$

Setup (2005-2012):
- **True emissions:** 10% annual growth rate
- **Prior emissions:** 2010 true emissions x random noises x 0.5, for all years

Hybrid approach reduce error (bias) by 50-74% (23-96%) compared to MB for SO$_2$

Joint inversion reduce error (bias) by by 3-18% (7-13%) compared to single species inversion for SO$_2$, except for 2005
Different magnitude and changing directions of NO$_2$ and SO$_2$ SCD from different retrieval products

- NO$_2$ NASA standard product is 50% smaller than DOMINO retrievals in densely populated and industrial regions.
- Posterior NO$_x$ emissions are more robust in Yangtze River Delta, Xinjiang, Ningxia, and Inner Mongolia.

(Qu et al., JGR, 2017)
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SO$_2$ SCD, GC – OMI, Jan 2010

- Consistent underestimate of volcanic SO$_2$ due to missing sources
- Inconsistent magnitude and signs of model-observation differences in mid latitude in NH, possibly caused by different cloud product and surface reflectivity
Discrepancies in different retrieval products can propagate into top-down emissions

Emissions in China

- Posterior NO\textsubscript{x} emissions from NASA SP is smaller than that from DOMINO by 39-46%
Discrepancies in different retrieval products can propagate into top-down emissions

- Posterior NO$_x$ emissions from NASA SP is smaller than that from DOMINO by 39-46%
- China’s posterior SO$_2$ emission from NASA SP is 17-46% smaller than top-down estimates from BIRA product.
Evaluation with in-situ measurement show better consistency of posterior concentration in China but worse in US

Surface SO$_2$ concentration in China (Jan 2010) [ug/m$^3$]
Evaluation with in-situ measurement show better consistency of posterior concentration in China but worse in US

Surface $SO_2$ concentration in China (Jan 2010) [ug/m$^3$]

- Posterior surface $SO_2$ concentration constrained by BIRA product reduce bias and improve correlation while SP posterior degrade the performance in China.
Evaluation with in-situ measurement show better consistency of posterior concentration in China but worse in US

Surface SO$_2$ concentration in US [ppbv]

- Posterior surface SO$_2$ concentration in US has worse correlation
- SP posterior has reduced bias compared to AQS measurements
Summary

• Reduced posterior emission error when assimilating NO₂ and SO₂ to optimize NOₓ and SO₂ emissions simultaneously

• Different NO₂ and SO₂ retrievals lead to ~ 50% discrepancies in posterior emissions

• Differences in SO₂ retrievals are possibly related to different cloud product and meteorology

• Improved consistency in posterior simulation of SO₂ concentration with surface measurements in China, but no obvious improvements in US
OMI SO2 SCD from different products, Jan 2010