

## Regional and global $\text{NO}_x$ and $\text{SO}_2$ emissions from different inversion frameworks and associated impacts on secondary pollutant estimates

Zhen Qu (1), Daven Henze (1), Jun Wang (2), Xiaoguang Xu (2), and Yi Wang (2)

(1) University of Colorado Boulder, Mechanical Engineering, United States (zhen.qu@colorado.edu), (2) University of Iowa, Iowa, United States

Accurate estimates of the magnitude and trends of emissions of nitrogen oxides ( $\text{NO}_x$ ) and sulfur dioxide ( $\text{SO}_2$ ) are important for improving understanding of air pollution and the effectiveness of emission control strategies. We estimate regional  $\text{NO}_x$  and  $\text{SO}_2$  emissions for China and the United States at the  $0.5^\circ \times 0.67^\circ$  resolution using newly derived mass balance and 4D-Var multi-species framework based on GEOS-Chem adjoint model, and extend a recently developed hybrid (mass-balance / 4D-Var) method to estimate long-term (2005-2012) emissions of these two species. In the multi-species framework, we optimize  $\text{NO}_x$  and  $\text{SO}_2$  emissions simultaneously, using both  $\text{NO}_2$  and  $\text{SO}_2$  observations from the OMI remote sensing instrument. Due to chemical interactions between these two species, inclusion of  $\text{SO}_2$  observations leads to  $\sim 30\%$  grid-cell level differences in posterior  $\text{NO}_x$  emissions compared to those constrained only by  $\text{NO}_2$  observations. In eight-year pseudo observation tests, posterior  $\text{NO}_x$  and  $\text{SO}_2$  emissions from 4D-Var multi-species framework have the smallest total normalized mean square error (NMSE, 0.75 – 5.09 for years from 2005 to 2012) compared to the true emissions. In other scenarios with prior emission being half of the true emission, posterior emissions in joint mass balance (NMSE = 0.86 – 10.82) and single species hybrid inversions (NMSE = 0.75 – 5.78) have smaller error than those in single species mass balance inversions (NMSE = 0.88 – 12.28).  $\text{NO}_x$  and  $\text{SO}_2$  emissions are also correlated through the amount of fuel combustions, since each of them is the product of species emission factors and activities. To incorporate this correlation into the inversion, we assimilate  $\text{NO}_2$  and  $\text{SO}_2$  observations to optimize seven sector-specific emission scaling factors. Posterior  $\text{NO}_x$  emissions from the sector-based inversion in US have the same seasonality as the prior, while seasonality in posterior emissions from species-based inversion is different. We also apply the hybrid approach to estimate global  $\text{NO}_x$  emissions from 2005 to 2015 at the  $2^\circ \times 2.5^\circ$  resolution. While top-down  $\text{NO}_x$  emissions in China started to decrease from 8.0 TgN/year in 2011 to 7.0 TgN/year in 2015, the reduction of top-down  $\text{NO}_x$  emissions in the United States has slowed down from 2010. No clear  $\text{NO}_x$  emission trend is observed in West Europe and Japan, where averaged top-down estimates are 1.2 TgN/year and 0.66 TgN/year during this eleven-year period. Comparing to simulations using HTAP bottom-up emissions in 2010, top-down constraints on  $\text{NO}_x$  emissions leads to grid cell level changes of  $-0.8$  –  $6.9$  ppbv  $\text{O}_3$  during the summer (mean 0.1 ppbv) and  $-4.2$  –  $0.9$   $\mu\text{g}/\text{m}^3$  annual mean  $\text{PM2.5}$  concentration (mean  $-1.3$   $\mu\text{g}/\text{m}^3$ ) over East Asia. From 2005 to 2012, the average summer  $\text{O}_3$  concentration in China increases by 0.5 ppbv ( $-6.4$  –  $7.2$  ppbv grid cell level changes) although decreases are observed around populated and industrial regions dominated by  $\text{NO}_x$  titration. Annual mean  $\text{PM2.5}$  concentration increases by 0.1  $\mu\text{g}/\text{m}^3$  ( $-4.6$  –  $8.6$   $\mu\text{g}/\text{m}^3$  grid level changes) during the same period. Long-term impacts on premature deaths will be further evaluated to quantify the impact of emission regulations.