



Regional and global NO_x and SO₂ emissions from different inversion frameworks and associated impacts on secondary pollutant estimates

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Accurate estimates of the magnitude and trends of emissions of nitrogen oxides (NO_x) and sulfur dioxide (SO₂) are important for improving understanding of air pollution and the effectiveness of emission control strategies. We estimate regional NO_x and SO₂ emissions for China and the United States at the 0.5° x 0.67° 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 NO_x and SO₂ emissions simultaneously, using both NO₂ and SO₂ observations from the OMI remote sensing instrument. Due to chemical interactions between these two species, inclusion of SO₂ observations leads to ~30% grid-cell level differences in posterior NO_x emissions compared to those constrained only by NO₂ observations. In eight-year pseudo observation tests, posterior NO_x and SO₂ 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). NO_x and SO₂ 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 NO₂ and SO₂ observations to optimize seven sector-specific emission scaling factors. Posterior 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 NO_x emissions from 2005 to 2015 at the 2° x 2.5° resolution. While top-down 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 NO_x emissions in the United States has slowed down from 2010. No clear 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 NO_x emissions leads to grid cell level changes of -0.8 – 6.9 ppbv O₃ during the summer (mean 0.1 ppbv) and -4.2 – 0.9 ug/m³ annual mean PM_{2.5} concentration (mean -1.3 ug/m³) over East Asia. From 2005 to 2012, the average summer O₃ 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 NO_x titration. Annual mean PM_{2.5} concentration increases by 0.1 ug/m³ (-4.6 – 8.6 ug/m³ grid level changes) during the same period. Long-term impacts on premature deaths will be further evaluated to quantify the impact of emission regulations.