



The Impacts of Fossil Fuel Emission Uncertainties and Accounting for 3-D CO₂ Chemical Production on Inversion for Natural Fluxes Using Satellite and In Situ Observations

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Atmospheric inversions for estimating natural carbon dioxide (CO₂) fluxes typically do not allow for adjustment of fossil fuel emissions. Given the significant spatial pattern differences among emission inventories and the usually incorrect specification of the vertical distribution of international bunker emissions or even complete omission of those emissions in inversions, important errors in inferred natural fluxes could potentially occur. Also, most inversions place CO₂ release from fossil fuel combustion and other sources entirely at the surface. However, a portion of fossil fuel and biospheric carbon emissions (~1 Pg C yr⁻¹) actually occurs in the form of reduced carbon species including carbon monoxide (CO) and volatile organic compounds (VOCs), which are eventually oxidized to CO₂ downwind of the emissions. As noted by a few previous studies, omission of this ‘chemical pump’ can result in a significant redistribution of the inferred fluxes among regions. In this study, we assess the impacts of different prescriptions of fossil fuel emissions and accounting for 3-D atmospheric CO₂ production on posterior natural fluxes during 2009-2010, with a novel aspect of considering satellite CO₂ observation-based as well as surface in situ-based inversions. We employ an established, relatively high-resolution, global, batch Bayesian synthesis inversion system based on the PCTM transport model. For baseline emissions, we use the ODIAC global fossil fuel CO₂ data product, which shares country-level estimates with the CDIAC gridded dataset but also provides international bunker emissions. We then apply period-specific 3-D CO loss rates archived from a state-of-the-art GEOS-5 chemistry and climate model simulation in a forward PCTM run to simulate the distribution of CO₂ originating from oxidation. To avoid double-counting, we subtract amounts from the surface emitted in the form of fossil and biospheric CO, CH₄, and non-methane VOCs. We find that the posterior large-scale fluxes are generally insensitive to the choice of fossil fuel inventory (CDIAC vs. ODIAC) and assumptions about international bunker emissions. Accounting for 3-D CO₂ production and the surface correction results in regional shifts in the global sink, e.g., from land to ocean in an inversion using GOSAT satellite data, with the magnitude and even sign of shifts dependent on assumptions about the surface correction. The GOSAT inversion is more sensitive to the chemical pump over land and ocean than an inversion using in situ observations, due to the differences in the horizontal and vertical sampling of the two observation types. Overall, the impact of 3-D CO₂ production appears to be minor relative to the differences between the in situ and GOSAT inversions.