Weight of Evidence for Significant Soil NO_x Emissions from Agricultural Systems in California

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Introduction

Nitrogen dioxide (NO_2) is a reactive gas with a daytime lifetime of about 5 hours with respect to its reaction with the hydroxyl radical [Laughner & Cohen, 2019] in the summertime and 20-25 hours during winter [Shah et al., 2020; Sun et al., 2021]. Nitrogen oxides ($NO_x = NO + NO_2$) serve as important precursors to tropospheric ozone (O_3) and fine particulate matter ($PM_{2.5}$) with consequent adverse effects including premature death [Caiazzo et al., 2013] cardiovascular mortality [Cohen et al., 2017], respiratory diseases [Meng et al, 2010], and agricultural productivity losses [Sampedro et al., 2020]. The primary sources of NO_x involve the thermogenic release during high-temperature combustion in air from vehicles [Tan et al., 2019] and power plants [de Foy et al., 2015], lightning [Schumann & Huntrieser, 2007], biomass burning [Campbell et al., 2022]. However, there are also microbial emissions from nitrification/denitrification processes in soils [Williams & Fehsenfeld, 1991; Yienger & Levy, 1995; Oikawa et al., 2015] that have been in global atmospheric chemistry models for decades. As anthropogenic emissins continue to decline due to regulatory efforts, the proportional impact of soil emissions is rising per force. In fact, a recent modeling study by Silvern et al. [2019] for the continental US (CONUS) estimates the proportion of total emissions from anthropogenic fossil fuel combustion to be only 42% in 2017 (compared to ~18% from soil emissions.)

Despite the long-term progress in NOx reductions, non-attainment of national air quality standards persists throughout California's Central Valley and other intensively agricultural areas [Parrish et al., 2017]. Trousdell et al. [2019] show that more than one in three days between May and October violates the National Ambient Air Quality Standards (NAAQS) for ground-level O_3 (70 ppb) in the Southern San Joaquin Valley (from 2006-2015). de Foy et al. [2020] also report that O_3 concentrations in the San Joaquin Valley (SJV) exceeded the NAAQS on 71 days in 2017 and 43 days in 2018. Similarly, Burley et al. [2016] found that the Sierra Nevada Mountains also routinely experience O_3 exceedances during the summer months. As substantial reductions in anthropogenic NO_x emissions have been achieved, recent work has demonstrated that the O_3 formation regime has shifted to predominantly NO_x-limited in these non-attainment inland areas since the 2010s [de Foy et al., 2020], meaning that the O_3 formation becomes more sensitive to misunderstanding of other non-traditional NO_x sources attributed to biogenic emissions [Silvern et al., 2019; Geddes et al., 2022].

Figure 1 shows the plateau behavior of both ozone and PM2.5 design values in two agricultural air basins, the San Joaquin Valley (SJVAB) and the Salton Sea (SSAB). Notice the absence of a decreasing trend in the last 5-6 years for both air quality parameters.

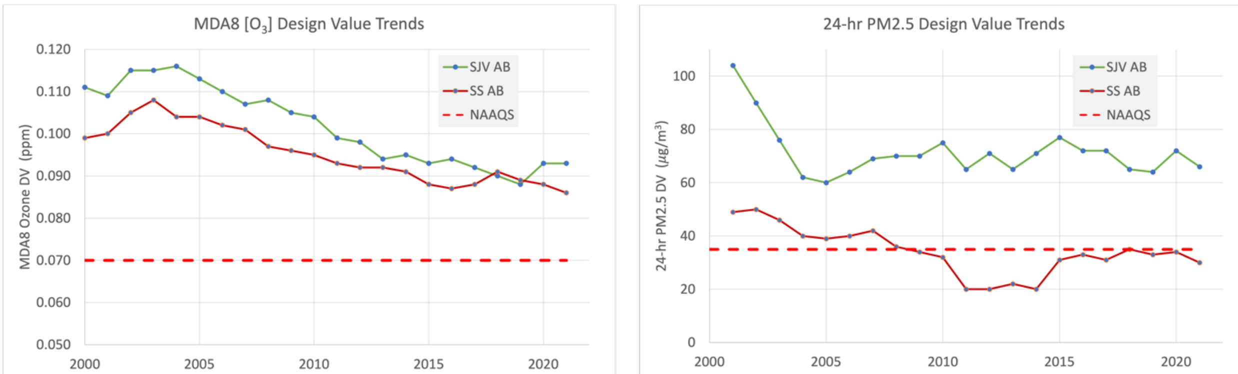


Figure 1. Trends in ozone and PM2.5 in the San Joaquin Valley and Salton Sea Air Basins over the 21st century. Data from CARB's iADAM: Air Quality Data Statistics web site.

Independent Lines of Evidence for Agricultural Soil Emissions

1. Oikawa et al. (2015). This study reported some of the highest soil NO_x emissions ever observed (up to 280 kg N ha^{-1} yr^{-1}) at a plot in Holtville, CA in the Imperial Valley. Their analysis indicated that the default soil NOx parameterization in their WRF-Chem model was ***at least an order of magnitude smaller*** than necessary to match the observations of NO_x. Their photochemical model responded to augmented soil NO_x by increasing average ozone between 2-8.5 ppb.

2. Parrish et al. (2017). Developing an empirical model to investigate the asymptotic behavior of ozone improvements in air basins all across southern California, this study found that the San Joaquin Valley and Salton Sea air basins exhibited a time evolution that differed substantial from all other air basins. Namely, their asymptotes were much larger than other air basins indicating the presence of sources that have not been decreasing under regulatory efforts like in the rest of California. The authors suggest this could be due to the intensive agricultural activities in these areas (whether from BVOC or soil NO_x precursor emissions.)

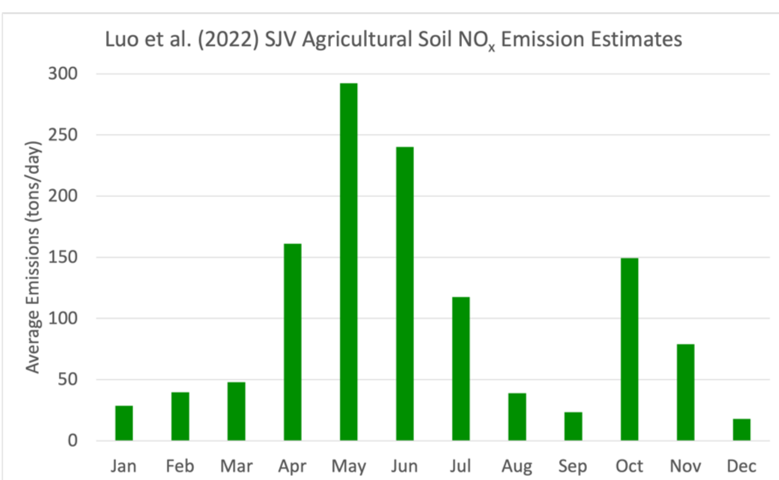
3. Almaraz et al. (2018). Based on a bottom-up soil N model and top-down aircraft measurements [Trousdell et al., 2019], the authors argue that agricultural soils emit 20-32% of the state's total NO_x (with another 5-9% coming from natural, unmanaged soils). The modeling study found the average NO_x flux out of croplands to be about 20 kg N ha^{-1} yr^{-1}. The airborne study [Trousdell et al., 2019] focused on the region surrounding Fresno, CA estimating a total emission rate of 215 $\pm$ 33 tons/day, while the sum from all counties in the flight domain for the CARB inventory (CEPAM) amounted to 104 tons/day. They also show that rural counties in the SJV indicate no decreases in near-surface O_3 in the period 2006-2016, in contrast to their urban counterparts.

4. Guo et al. (2020). By coupling the DeNitrification-DeComposition (DNDC) biogeochemical model with CMAQ they concluded that soil emissions contribute only 1.1% of total anthropogenic NO_x emissions in California. The authors do admit to finding some large emission regions in the warm Imperial Valley, suggesting that the default temperature coefficient for nitrification (Q_{10}) may need to be revised, but left it unchanged for this study.

5. Wang et al. (2021). These researchers used a much stronger, observation-based temperature response of soil NO_x emissions, which was about 30% larger at 30°C but 300% larger at 40°C. Using this high-temperature adjustment to the scheme they found improved linear correlations between GEOS-Chem and satellite column NO_2 observations. Such a change in parameterization would significantly enhance emissions from what are considered the hotspots in the state, the San Joaquin and Imperial Valleys, due to their intensive N fertilization and warm climate.

6. Sha et al. (2021). These researchers made some modest changes to the soil NO_x parameterization in WRF-Chem (Berkeley Dalhousie Iowa Soil NO Parameterization, BDISNP). Changes included better representation of land cover distributions, soil temperature, and emission pulses, as well as including fertilizer N emissions from agricultural soils. For the one month they modeled (July 2018) the researchers estimated that soil emissions account for ~40% of California's total NO_x emissions, significantly increasing the surface O_3 (+23%) concentrations in rural California.

7. Luo et al. (2022). Using an updated nitrogen scheme in an agroecosystem model (Fertilizer Emission Scenario Tool for the CMAQ, FEST- C) the researchers simulated the reactive N emissions from fertilized soils across the contiguous United States. This team reported soil NO_x emissions by county, showing annual average emissions from the 8 counties of the San Joaquin Valley Air Basin to be estimated at ***100 tons/day***. For the sake of comparison, recent CARB estimates of all anthropogenic NO_x emissions in the air basin amount to about 200 tons/day. As for the seasonal distribution of such emissions, their model results are shown in Figure 2 for the SJV air basin.



8. Wang et al. (2023). Using OMI satellite NO_2 data from 2009-2020, the authors show patterns over cropland areas that indicate a soil-like T and soil moisture dependence, and they show the absence of a change over croplands (0.0 ± 0.4 %a^{-1}) as opposed to continued decreases (-3.7 ± 0.3 %a^{-1}) over urban areas.

The combined weight of evidence, therefore, indicates that soil NO_x emissions from agricultural soils are most probably influencing the persistent air quality problems in rural California.

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