Integrated Assessment of the Effects of  $NH_3$ ,  $NO_2$ , PM,  $SO_2$ , and VOC Emissions on  $O_3$  and  $PM_{2.5}$  Concentrations and Trends in New York State

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## Abstract

Large ( $^{50}$  – 85%) reductions in anthropogenic emissions occurred in the northeastern U.S. and eastern Canada between 1995 and 2015, providing an opportunity to apply integrated approaches to link (1) emissions with ambient air pollutant concentrations in New York State, and (2) primary with secondary air pollutants. Moderate responses of ozone ( $O_3$ ) and  $PM_{2.5}$  elemental carbon (EC) and organic carbon ( $O_3$ ), together with limitations and uncertainties in emission estimates and ambient measurements, highlight the need for further study.

Multi-site mean annual  $SO_2$  PM<sub>2.5</sub> sulfate ( $SO_4$ ) concentrations tracked regional (multi-state and province) and New York State  $SO_2$  emissions between 1999 and 2015. Results indicate that on average 0.5  $\mu$ g m<sup>-3</sup>  $SO_4$  originated from natural emissions or from outside the region.

Reductions of anthropogenic CO and VOC emissions led to corresponding decreases in ambient CO and VOC species concentrations. Multi-site mean annual nitrogen dioxide ( $NO_2$ ) concentrations, mean annual  $PM_{2.5}$  nitrate ( $NO_3$ ) concentrations, and total (wet + dry) annual nitrate deposition tracked regional  $NO_x$  emission reductions.

Between 1990 and 2015,  $O_3$  maxima (annual 4<sup>th</sup>-highest peak daily 8-hour  $O_3$  mixing ratio) declined from multi-site averages exceeding 80 ppbv to averages below 70 ppbv, but did not follow regional  $NO_x$  emission trends as closely as either  $NO_2$  or PM  $NO_3$  did. By 2014, the highest summer  $O_3$  maxima at rural sites were lower than spring maxima, due to a decrease in summer maxima. New York City sites continued to show higher summer than spring mean daily peak 8-hour  $O_3$  mixing ratios and exhibited increases during cooler months.

PM<sub>2.5</sub> EC and OC trends occurred at some sites but are not yet evident at others. A consistent slope of annual-average OC-vs-EC is maintained across all sites except Bronx (BRX), with statistically-significant (p < 0.0001) intercept and slope of OC =  $0.86 + 1.66 \times EC$ ,  $r^2 = 0.90$ . This spatial and temporal coherence implies that most sites are influenced by similar emission source types, and have responded similarly to reductions of primary PM<sub>2.5</sub> emissions or combustion-related precursors of secondary organic aerosol. The lowest mean annual OC (~1  $\mu$ g m<sup>-3</sup>) and EC (~0.2  $\mu$ g m<sup>-3</sup>) concentrations are recorded at Whiteface Mountain Base, possibly representing values that are close to regional background.