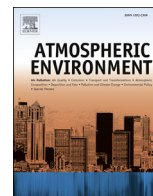




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# Long-term NO<sub>x</sub> trends over large cities in the United States during the great recession: Comparison of satellite retrievals, ground observations, and emission inventories



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## HIGHLIGHTS

- Derived multi-year urban NO<sub>x</sub> trend from satellite (OMI) and ground observations (AQS).
- Revealed NO<sub>x</sub> responses to the 2008 Economic Recession by OMI and AQS.
- The trend not well captured by emissions used for national air quality forecasting.
- Demonstrated how to use space and ground observations to evaluate emission updates.

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## ABSTRACT

National emission inventories (NEIs) take years to assemble, but they can become outdated quickly, especially for time-sensitive applications such as air quality forecasting. This study compares multi-year NO<sub>x</sub> trends derived from satellite and ground observations and uses these data to evaluate the updates of NO<sub>x</sub> emission data by the US National Air Quality Forecast Capability (NAQFC) for next-day ozone prediction during the 2008 Global Economic Recession. Over the eight large US cities examined here, both the Ozone Monitoring Instrument (OMI) and the Air Quality System (AQS) detect substantial downward trends from 2005 to 2012, with a seven-year total of –35% according to OMI and –38% according to AQS. The NO<sub>x</sub> emission projection adopted by NAQFC tends to be in the right direction, but at a slower reduction rate (–25% from 2005 to 2012), due likely to the unaccounted effects of the 2008 economic recession. Both OMI and AQS datasets display distinct emission reduction rates before, during, and after the 2008 global recession in some cities, but the detailed changing rates are not consistent across the OMI and AQS data. Our findings demonstrate the feasibility of using space and ground observations to evaluate major updates of emission inventories objectively. The combination of satellite, ground observations, and in-situ measurements (such as emission monitoring in power plants) is likely to provide more reliable estimates of NO<sub>x</sub> emission and its trend, which is an issue of increasing importance as many urban areas in the US are transitioning to NO<sub>x</sub>-sensitive chemical regimes by continuous emission reductions.

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## 1. Introduction

Nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>) are key precursors to tropospheric ambient ozone (O<sub>3</sub>) and fine particulate matter (PM<sub>2.5</sub>) (Crutzen and Gidel, 1983; Spicer, 1983), which has been

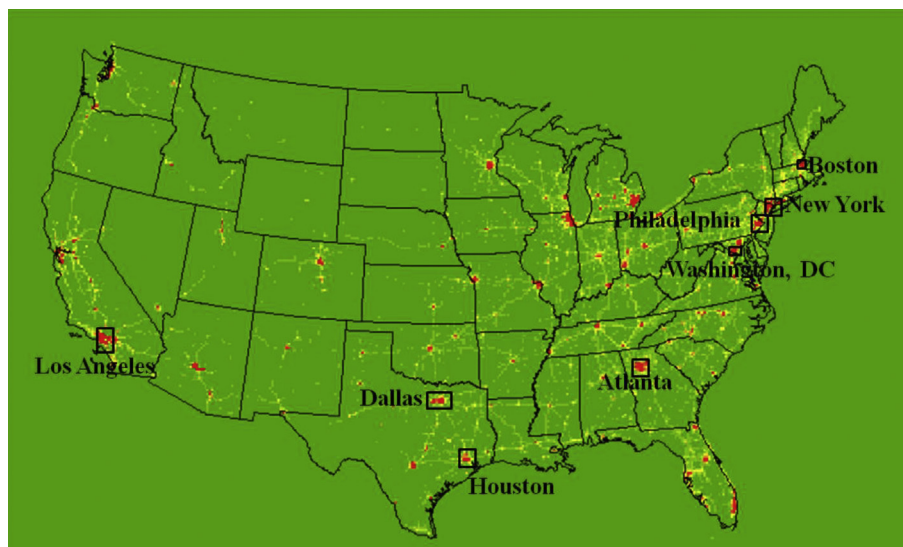
associated with adverse health effects, including respiratory diseases and cardiovascular mortality (Pope et al., 2002; Jerrett et al., 2009). As of December 2013, the United States Environmental Protection Agency (US EPA) estimates that more than one-third of the US population lives in areas that exceed the national ambient air quality standards (NAAQS) for either  $O_3$  or  $PM_{2.5}$  (US EPA, 2014a, 2014b). To assist state and local agencies in mitigating the effects of unhealthy levels of air pollution, the National Air Quality Forecasting Capability (NAQFC) system (Otte et al., 2005), currently operated by the National Weather Service, was designed to provide air quality forecasting guidance over the contiguous United States, Hawaii, and Alaska for next day forecasts (Stajner et al., 2012).  $NO_x$  are emitted from both anthropogenic sources (transportation, power plants, and fertilizers) and natural sources (biomass burning, lightning, and soils) (Warneck, 2000). Once emitted,  $NO_x$  reacts with volatile organic compounds under sunlight to form tropospheric ozone (Liu et al., 1987) and particulate nitrate, an important component of  $PM_{2.5}$ . Hence, quantifying the amount of  $NO_x$  emitted into the atmosphere is essential for reliable prediction of surface ozone and  $PM_{2.5}$ .

It is often challenging to provide accurate estimates of  $NO_x$  emissions for time-sensitive applications such as NAQFC, given the rapid progression of emission control and other socioeconomic events that affect emission loading (e.g., Harley et al., 2005; van der A et al., 2008; Stavrakou et al., 2008; Konovalov et al., 2010; Pinder et al., 2011; Castellanos and Boersma, 2012; Duncan et al., 2013). NAQFC relies on national emission inventories (NEIs) to account for thousands of anthropogenic emission sources and other emission models for natural sources. The substantial cost and effort entailed in collecting data relevant for compiling NEIs are prohibitive for frequent and timely updates of  $NO_x$  NEIs using conventional emission modeling approaches. As a result, the emission data used in NAQFC are several years behind the forecasting year, imposing uncertainties on air quality forecasting (Tong et al., 2012).

Can satellite-based emission data provide reliable information in order to rapidly update  $NO_x$  emission inventories and thereby support NAQFC-type air quality applications?  $NO_2$  retrievals from polar orbiting satellite sensors such as SCIAMACHY and GOME have been used to update anthropogenic  $NO_x$  emission inventories (Martin et al., 2003; Lamsal et al., 2011; Mijling and van der A,

2012). This satellite-based approach, while showing great potential to reduce the emission time lag, has yet to be verified with independent data sources. To evaluate the robustness of this approach, this study thus compares the long-term  $NO_2$  trends derived from the Ozone Monitoring Instrument (OMI) (Boersma et al., 2007) with the operational emission data used in NAQFC predictions, with emphasis on both the emission changes between major updates and the year-to-year progression over eight metropolitan areas from 2005 to 2012. A third data source, the EPA's AQS ground observations, serves as an independent reference to help verify urban  $NO_x$  trends. Estimation of future year emissions for air quality and climate modeling often relies on emission projections (Bond et al., 2004; Granier et al., 2011). NAQFC emissions are updated annually based on the available emission inventories, emission measurements, and projections. Rigorous evaluations of these annual updates, however, have not been performed regularly, largely because of the lack of observational data to directly verify emission projections. Meanwhile, AQS data have been used as a proxy for urban emissions since morning rush-hour concentrations are predominantly influenced by emissions from heavy commuter traffic (Godowitch et al., 2010). The AQS measurements have been coupled with other data to identify source strength and the origin of reactive nitrogen oxides over the Southeastern United States (Tong et al., 2005). Therefore, all three data sources provide independent quantification of  $NO_x$  emissions that can be compared over a fixed time period.

The intercomparisons of changes in  $NO_x$  from NAQFC, OMI, and AQS are expected to serve dual purposes: 1) to compare the  $NO_x$  trends derived from space and ground observations; and 2) to evaluate NAQFC emission updates against satellite and ground observations. We focus here on the  $NO_x$  trends over eight major metropolitan areas (Fig. 1) where  $NO_x$  emission density is high and AQS monitors are abundant. We further concentrate on the  $NO_x$  trends during summertime when the  $NO_x$  photochemical lifetime is shorter (4 h at noon), the carryover from the previous day is limited and regional transport is at minimal, yielding a column that is especially representative of local surface emissions (Russell et al., 2010). Further, the number of available satellite measurements is maximized during summer months, when cloud cover is lowest. Finally, these studied areas are among the most populous cities in



**Fig. 1.** Locations of the eight selected metropolitan statistical areas, which are among the most densely populated cities in the United States. The background color represents  $NO_x$  emission density based on the NAQFC emission data, with red indicating high  $NO_x$  emission density. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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