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Constraining NO_x emissions using satellite NO_2 measurements during 2013 DISCOVER-AQ Texas campaign

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HIGHLIGHTS

- Constraining anthropogenic and biogenic NO_x emissions using OMI.
- A large decrease (30–60%) in anthropogenic emissions in urban areas.
- An increase (52%) in soil-biogenic emissions in rural regions.
- Improvements of simulating NO_2 levels using the constrained emission inventory.

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ABSTRACT

Reliable emission inventories are key to precisely model air pollutant concentrations. The relatively large reduction in NO_x emissions that is well corroborated by satellite and in-situ observations over southeast Texas has resulted in discrepancies between observations and regional model simulations based on the National Emission Inventory (NEI) provided every three years in U.S. In this study, a Bayesian inversion of OMI tropospheric NO_2 is conducted to update anthropogenic sources of NEI-2011 and soil-biogenic sources from BEIS3 (Biogenic Emission Inventory System version 3) over southeast Texas and west Louisiana during the 2013 DISCOVER-AQ Texas campaign. Results reveal that influences of the a priori profile used in OMI NO_2 retrieval play a significant role in inconsistencies between model and satellite observations, which should be mitigated. A posteriori emissions are produced using the regional Community Multiscale Air Quality (CMAQ) model associated with Decoupled Direct Method (DDM) sensitivity analysis. The inverse estimate suggests a reduction in area (44%), mobile (30%), and point sources (60%) in high NO_x areas (ENO_x> 0.2 mol/s), and an increase in soil (~52%) and area emissions (37%) in low NO_x regions (ENO_x< 0.02 mol/s). The reductions in anthropogenic sources in high NO_x regions are attributed to both uncertainty of the priori and emissions policies, while increases in area and soil-biogenic emissions more likely resulted from under-estimation of ships emissions, and the Yienger-Levy scheme used in BEIS respectively. In order to validate the accuracy of updated NO_x emissions, CMAQ simulation was performed and results were evaluated with independent surface NO_2 measurements. Comparing to surface monitoring sites, we find improvements (before and after inverse modeling) for MB (1.95, −0.30 ppbv), MAB (3.65, 2.60 ppbv), RMSE (6.13, 4.37 ppbv), correlation (0.68, 0.69), and IOA (0.76, 0.82). The largest improvement is seen for morning time surface NO_2 for which RMSE and MB are reduced considerably by ~60% (5.05 to 2.07 ppbv) and 109% (3.49 to −0.3 ppbv). However, underprediction of model NO_2 in Houston on 09/25–09/26 likely resulting from unprecedented local NO_x sources from the Houston Ship Channel (HSC) becomes more evident. Overall, results show that use of OMI tropospheric NO_2 columns can substantially reduce the uncertainty of bottom-up emissions for a regional study which should be extended to larger domains (e.g. entire U.S. or North America).

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1. Introduction

Nitrogen oxides ($NO_x = NO + NO_2$) emissions produced from combustion of fossil fuels (Noxon, 1978), biomass burning (van der Werf et al., 2006), soil microbial activity (Yienger and Levy, 1995) and lightning (Choi et al., 2009), are not only implicated in formation of tropospheric ozone and acid rain, but also are known as one of the main criteria air pollutants, harming human respiratory systems (Kampa and Castanas, 2008). Promisingly, control technologies and strategies implemented by government regulatory agencies have succeeded in reducing anthropogenic NO_x emissions in the U.S. According to EPA's National Emissions Inventory (NEI) air pollutant emissions trends data (http://www.epa.gov/ttnchie1/trends/), anthropogenic sources of NO_x from both stationary and mobile sources declined by 45% in the U.S. from 2000 to 2014. Chemical Transport Models (CTMs) to some degree have managed to simulate temporal and spatial variations in ozone (Li et al., 2015), VOC (Pan et al., 2015), PM (Jeon et al., 2015), HONO (Diao et al., 2016) and acid deposition (Butler et al., 2005) on the basis of bottom-up NO_x emission inventories, even though regional model simulations (e.g., Community Multiscale Air Quality (CMAQ) and Weather Research and Forecasting (WRF)-Chem models) are still problematic in simulating NO_x concentrations (e.g., Kim et al., 2011; Choi et al., 2012; Choi, 2014; Choi and Souri, 2015a). This could be attributed to uncertainties of NO_x emissions, chemical mechanisms or depositions. Uncertainty of NO_x emissions is of particular scientific interest because bottom-up emissions are susceptible to rapid obsolescence due to both rapid anthropogenic emissions reductions, and fast response of NO_2 to its emissions resulting from the relatively short life time of NO_2 (~3 h–2 days; Martin et al., 2003). This highlights specific limitations of bottom-up emission inventories provided every 3 years for the U.S., biasing results of air quality simulation for the present study. In a previous study (Choi and Souri, 2015a), we investigated changes in anthropogenic emissions and their impacts on ozone using a regional model (WRF-Chem) employing two different scenarios for September 2013 in Texas. The first scenario using NEI-2005 showed that the model substantially over-predicted surface NO_2 by a factor of ~3.6 over Texas. In the second scenario in Choi and Souri (2015a), using NEI-2011 emissions, this over-estimation significantly decreased reaching a factor of 1.7; but it was still noticeable underscoring the uncertainties of NO_x emission in the region.

One way to improve the emission inventory is to use observations to constrain emissions, that is, to infer what must have been emitted in order to observe measured ambient values. Inverse modeling of sources of atmospheric trace gases is a well-established technique that incorporates this inference process and has been addressed in many studies (e.g., Martin et al., 2003; Shim et al., 2005; Zhang et al., 2008; Chai et al., 2009; Gu et al., 2014; Huang et al., 2014). These studies belong to a large domain of chemical data assimilation. In this procedure, emissions are optimized in order to reduce differences between modeled and observed data. The differences between inverse modeling studies can be characterized by the type of observations (e.g., surface, aircraft and satellite remote sensing), the way observations are related to emissions (e.g., direct mapping, brute force, Decoupled Direct Method (DDM) and adjoint), and how the optimum estimation is guaranteed (e.g., ordinary least squares, Bayesian theorem and Kalman filter) given the errors of observations and emissions (Sandu and Chai, 2011).

Updating emission inventories using satellite measurements is a common application for which remote measurements are effective (Streets et al., 2013 and the references therein). Due to spatial inhomogeneity in distributions of NO_x concentrations and limited monitoring sites, satellite remote sensing of tropospheric NO_2 is widely used for this purpose. The biggest advantage of using satellite data is the high spatial coverage that makes it possible to update emission inventories over large spatial domains. Instead, the large number of pre-processing tasks (e.g., removing a priori profile influences, aerosol and cloud effects, etc.), variety of sensors and retrieval methods might result in different answers from case to case (e.g., Kemball-Cook et al., 2015). Thus, the level of accuracy of retrievals and independent measurements are required to validate the a posteriori emission inventory.

Southeast Texas is home to large petrochemical industry which produces a considerable amount of anthropogenic VOC and NO_x emissions, often leading to ozone formation. The complexity of meteorological conditions in this coastal urban area (Souri et al., 2016; Li et al., 2015), as well as uncertainties of VOC and NO_x emissions (Choi and Souri, 2015a; Pan et al., 2015), have created challenges to providing consistent predictions of atmospheric chemistry. The present study is motivated by the opportunity to explore the possibility of using tropospheric NO_2 columns from Ozone Monitoring Instrument (OMI) aboard the Aura satellite to improve the most recent national emission inventory (NEI-2011) as well as soil-biogenic emissions from BEIS3 (Biogenic Emission Inventory System version 3) in southeast Texas. To end this, we use CMAQ version 5.0.2 with DDM (Cohan et al., 2005) to derive sensitivities of NO_2 levels to NO_x emissions; subsequently a Bayesian inversion method is used to update four NO_x emissions source categories (i.e., area, mobile, point and soil-biogenic). These form the basis for a new improved emissions inventory which we call NEI-2011n.

2. Measurements, modeling and method

2.1. Measurements

2.1.1. Air quality monitoring sites

Surface observational data consist of regular measurements from the Continuous Ambient Monitoring Stations (CAMS) and Air Quality System (AQS) sites operated by the TCEQ and EPA, respectively. The CAMS measurement network collects real-time meteorology and air pollutant concentrations. Measured parameters differ from station to station. Station density in southeast Texas is relatively high. The number of sites having meteorological and NO_2 measurements were 63 and 31 respectively in the 4-km modeling

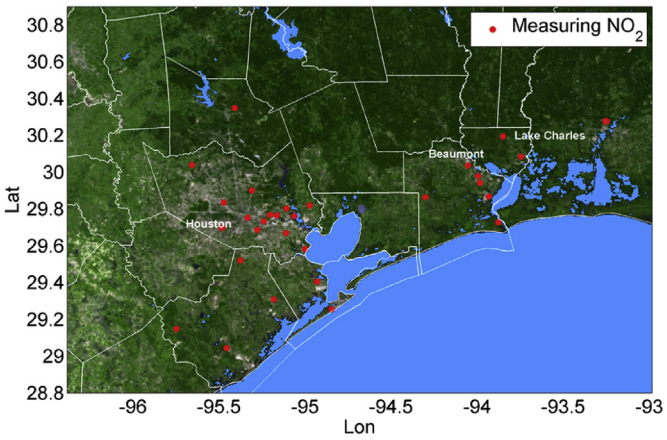

Fig. 1. Locations of CAMS and AQS NO_2 sites available in 2013 DISCOVER-AQ in our 4-km modeling domain. We refined NO_2 concentrations by taking account of other reactive nitrogen gases from model simulation. Three main metropolitan areas in this region are Houston-Galveston-Brazoria, TX, Beaumont-Port Arthur, TX and Lake Charles, LA.