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Assessment of the magnitude and recent trends in satellite-derived ground-level nitrogen dioxide over North America

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HIGHLIGHTS

- Trends in OMI-derived surface NO_2 are significantly correlated with in situ.
- Absolute concentrations and trend from in situ are twice those from satellite.
- This representativeness difference arises from spatial averaging vs. point values.
- In situ monitors tend to be in locations with elevated NO_2.
- Satellites offer additional information in regions without monitors.

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ABSTRACT

We estimate ground-level nitrogen dioxide (NO_2) concentrations from the OMI (Ozone Monitoring Instrument) over North America for the period 2005–2012. A chemical transport model (GEOS-Chem) is used to account for effects of the NO_2 profile on the column retrieval, and to relate OMI NO_2 columns to ground-level concentrations. The magnitude of the period-mean OMI-derived NO_2 concentrations is evaluated versus in situ measurements from air quality networks yielding a significant spatial correlation ($r = 0.81$) but OMI-derived values are lower with a slope of 0.4. Comparison of the in situ concentrations versus spatially resolved concentrations estimated from land use regression models reveals that this difference partially arises from representativeness difference due to preferential placement of in situ monitors at locations with enhanced NO_2, coupled with the OMI horizontal resolution. In situ observations provide information about local concentrations while OMI offers area-averaged information. The remaining difference is less readily explained and appears to include a combination of the effects of local unresolved geophysical processes affecting both the NO_2 retrieval and the vertical profile used to relate the column to ground level. We also evaluate trends over North America from OMI and in situ measurements for the period of 2005–2012. OMI derived ground-level NO_2 well reproduces the spatial pattern of the in situ trends ($r = 0.77$) and the slope of 0.4 versus the trend from in situ monitors is consistent with the slope versus mean concentrations. Absolute regional trends inferred from in situ measurements alone may overestimate area average changes. Nonetheless coincidently sampled ground-level NO_2 concentrations from OMI and in situ measurements for 2005–2012 exhibit similar relative decreases over Eastern (-6.5 ± 2.0%/yr, -7.1 ± 1.3%/yr), Western (-4.5 ± 1.1%/yr, -6.5 ± 0.7%/yr) and Central (-3.3 ± 2.3%/yr, -4.1 ± 0.8%/yr) North America.

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1. Introduction

Nitrogen oxides (NO_x = NO + NO_2) play a central role in tropospheric chemistry with implications for air quality, climate, and ecosystems. Nitrogen dioxide (NO_2) is a general marker of combustion, and in urban areas an indicator of traffic-related air pollution that is associated with premature mortality (Burnett et al., 2004; Stieb et al., 2002; Shin et al., 2008; Crouse et al., 2015), and with adverse effects on respiratory health (Filleul et al., 2005; Gauderman et al., 2005). NO_2 concentrations also have been used as an air pollution marker to assess patterns of environmental injustice and inequality (Clark et al., 2014). Outdoor air pollution is a leading environmental risk factor for premature mortality (Lim et al., 2012). Accurate observations of NO_2 concentrations and trends from satellite offer a valuable data source to understand ground-level air quality, which complements satellite based estimates of $PM_{2.5}$ (Cooper et al., 2012).

Satellite observations of trends in NO_2 have attracted considerable attention in recent years. Richter et al. (2005) used GOME and SCIAMACHY during 1996–2004 and reported a substantial decrease in tropospheric NO_2 columns over Europe and the United States, but a significant increase of ~50% over the industrial areas of China. Schneider and van der A (2012) analyzed nine-year (2002–2011) SCIAMACHY datasets and found decreases in tropospheric NO_2 over Europe and the US, and strong increase over China and many megacities in Asia. Hilboll et al. (2013) analyzed the long term trend in tropospheric NO_2 columns using multiple satellite observations over megacities. Geddes et al. (2015) examined the long-term trend in global ground-level NO_2 concentrations. Russell et al. (2012) used OMI for the period of 2005–2011 and observed a consistent decrease in tropospheric NO_2 in cities across the US, with an average total reduction of 32 ± 7%. Duncan et al. (2013) reported a clear scalar response of OMI NO_2 column data to NO_x emission reductions from power plants associated with the implementation of mandated emission control devices (ECDs) over United States for the period of 2005–2011.

Globally, few in situ NO_2 monitoring networks exist worldwide with sufficient measurements to assess long-term trends in ground-level NO_2. The United States Environmental Protection Agency observed nationally 33% decreases in annual mean ground-level NO_2 concentrations for the period of 2001–2010 (http://www.epa.gov/airtrends/2011/report/fullreport.pdf) that is attributed to NO_x emissions reductions from vehicles and electric utilities (McDonald et al., 2012). In Canada, Environment Canada's National Air Pollution Surveillance Program (NAPS) network detected a 41% decrease in annual average ground-level NO_2 concentrations from 1997 to 2011 (http://www.ec.gc.ca/indicateurs-indicators/default.asp?lang=en&n=C8BFC3F2-1). Recently Crouse et al. (2015) described long-term decreasing trends in NO_2 concentrations from NAPS stations in 10 Canadian cities during 1984–2009. Guerreiro et al. (2010) analyzed trends in ground-level NO_2 over different regions of Europe using in situ measurements during 1999–2008 and found a general decrease in most locations. Information on trends in ground-level NO_2 is limited elsewhere.

Ground-level NO_2 concentrations inferred from satellite observations have been evaluated with in situ surface measurements (e.g. Bechle et al., 2013; Boersma et al., 2009; Lamsal et al., 2008; Lee et al., 2011; McLinden et al., 2014; Wang and Chen, 2013). Ground-level NO_2 derived from OMI correlates well with in situ measurements however, OMI-derived ground-level NO_2 concentrations are underestimated compared to the in situ surface measurements (Wang and Chen, 2013; Lamsal et al., 2008). This underestimation of ground based monitoring networks may arise from systematic placement of monitors in regions with elevated concentrations (Loperfido and Guttorp, 2008), compared to area averages estimated by satellite observations at a spatial scale of more than 100 km^2.

Land Use Regression (LUR) is a widely used technique for predicting ambient air pollutant concentrations at high spatial resolution (Hoek et al., 2008). Typically, a large number (~100) of passive NO_2 monitors are deployed for several weeks to measure the relation of NO_2 with LUR variables such as land use and traffic counts. These input data are used to predict the spatial distribution of the pollutants in a specific region. This approach has been applied to determine the intraurban variation in ambient NO_2 concentrations for many North American cities (e.g. Jerrett et al., 2007; Henderson et al., 2007; Wang et al., 2013) and beyond (e.g. Hoek et al., 2015; Vienneau et al., 2013).

In the present study we determine the ground-level NO_2 from OMI over North America for the time period of 2005–2012 and use LUR datasets to understand the comparison with measurements from air quality monitoring networks over North America. Section 2 describes the OMI retrievals, GEOS-Chem model, and monitoring networks. In Section 3 we present the OMI derived ground-level NO_2 evaluation. This evaluation is interpreted in the context of LUR in Section 4. Section 5 discusses the time series analysis.

2. Data sets & model

2.1. Ground-level NO_2 estimation from the OMI satellite instrument

OMI is a nadir-viewing UV–visible spectrometer on board the Aura satellite that was launched in July 2004 and flies as part of the NASA A-train constellation (Levelt et al., 2006). The Aura satellite overpasses the equator at 13:40 local time in sun-synchronous ascending polar orbit. OMI provides daily global coverage of aerosols and trace gases, including NO_2, with a variable ground spatial resolution of 13 km × 24 km at nadir to 140 km × 26 km at swath edge. The mean OMI spatial resolution is 310 km^2. We use here the tropospheric NO_2 column data products available from NASA (Version 2.1, collection 3) (Bucsela et al., 2013; Lamsal et al., 2014). In brief, the tropospheric NO_2 column is derived with the following three steps, 1) NO_2 slant column determination using DOAS algorithm in the 405–465 nm wavelength range, 2) stratospheric and tropospheric NO_2 separation (Bucsela et al., 2013; Lamsal et al., 2014) and 3) air mass factor calculation by integrating the NO_2 relative vertical distribution (shape factors) weighted by altitude and cloud dependent scattering weight factors to convert the slant columns into NO_2 vertical columns (Palmer et al., 2001; Martin et al., 2002). We also consider data from the BEHR (Berkeley High-Resolution) product that is designed to resolve spatial features at high resolution (Russell et al., 2011). We exclude the cross-track pixels affected by row anomaly (http://www.knmi.nl/omi/research/product/rowanomaly-background.php), which was first noticed in the data in June 2007. We use OMI NO_2 columns with cloud radiance fraction <0.3, and solar zenith angles <78°.

We estimate ground-level NO_2 concentration from OMI for comparison with in situ measurements over North America. We follow the approach used by Lamsal et al. (2008) that combines information on the simulated NO_2 vertical profile with tropospheric NO_2 column from the satellite observation, and the spatial variation of NO_2 concentrations in the boundary layer. The GEOS-Chem NO_2 vertical profiles are taken coincident with OMI observations. The sub-pixel ground-level NO_2 concentration is estimated from the OMI tropospheric NO_2 column following the method of Lamsal et al. (2008, 2013) that determines an inhomogeneity factor from the satellite columns using the assumption of a well-mixed free troposphere within the grid box. The ground-level NO_2 mixing ratio S is estimated from the local OMI tropospheric NO_2 column Ω as: