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Investigating ambient ozone formation regimes in neighboring cities of shale plays in the Northeast United States using photochemical modeling and satellite retrievals

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HIGHLIGHTS

- We investigate ambient ozone formation regimes for cities adjacent to shale plays.
- Column densities of precursors retrieved from OMI and GOME-2 are different.
- NO_x emission controls would reduce ozone levels in most cities in the Northeast U.S.
- Ozone formation in New York was limited by VOC emissions in 2007–2009 and 2014.
- Shale-related emission controls have the potential to reduce urban ozone levels.

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ABSTRACT

This study investigates long-term (i.e., 2007–2014) fluctuations in ambient ozone formation regimes for cities adjacent to shale plays in the Northeast United States (U.S.). Ozone air quality in many cities of the Northeast U.S. does not meet the U.S. National Ambient Air Quality Standards (NAAQS), and understanding ambient ozone formation regimes is essential to develop effective air pollution mitigation strategies for cities violating the air quality standards. Since 2013, the U.S. has become the world's largest producer of tight oil and natural gas from shale rock, and previous studies show that emissions of air pollutant precursors from shale oil and gas-related activities would have the potential to affect ambient ozone air quality in adjacent cities of shale plays. This work leveraged (1) satellite-retrieved column densities of formaldehyde (HCHO) and nitrogen dioxide (NO2) from multiple instruments (i.e., Ozone Monitoring Instrument (OMI) and Global Ozone Monitoring Experiment-2 (GOME-2)); (2) photochemical air quality modeling and sensitivity analysis; and (3) ratios of satellite-retrieved air pollutant column densities to investigate ambient ozone formation regimes in neighboring cities of shale plays (i.e., Marcellus Shale) in the Northeast U.S. from 2007 to 2014. Our results show that ambient ozone formation in Boston, Pittsburgh, Philadelphia and Washington, D.C. (which are close to Marcellus Shale) was in the NOx -limited or transition regime during the period of study. Ambient ozone formation in New York City was in the transition regime during 2010–2013 and VOC -limited regime during 2007–2009 and in 2014. Based on the result of this study, we conclude that controls NOx emissions would mitigate ozone air pollution from 2007 to 2014 in most of the cities examined in this study. Controls of local VOC emissions would ease ozone air pollution in New York City during the study period. With projected increases in oil and gas production from shale plays in the Northeast U.S., air pollutant emissions from oil and gas-related activities are expected to increase in the future. The results of this study imply that controls of

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ozone precursor emissions from shale oil and gas-related activities could be a potential strategy for reducing ambient ozone formation in cities adjacent to the shale plays in Northeast U.S. in the future.
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1. Introduction

Ambient ozone formation is driven by complex nonlinear photochemistry of nitrogen oxides (NO_x), volatile organic compounds (VOC), and other species and can be considered a "NO_x-limited" or "VOC-limited" regime (Carter, 1994; Sillman, 1999). The NO_x-limited and VOC-limited regimes mean ambient ozone formation is mainly controlled by emissions of NO_x and VOCs, respectively (Sillman, 1999). Reductions in NO_x emissions from local and/or upwind sources will decrease ambient ozone formation (and ground-level ozone concentrations) in NO_x-limited areas but increase ozone formation in VOC-limited areas. On the other hand, controls of VOC emissions will decrease ozone formation in areas with the VOC-limited regime but increase ozone formation in NO_x-limited areas. Otherwise, both reductions in NO_x and VOC emissions will decrease ozone formation in the transition regime. Since ozone air pollution mitigation is a complex issue, understanding ambient ozone formation regimes is essential to developing regional air pollution mitigation strategies to comply with air quality standards. Downey et al. (2015) conducted regional air quality modeling and sensitivity analyses to examine how ambient ozone concentrations in urban areas would respond to controls of anthropogenic NOx and VOC emissions in the U.S. The results of their study show at least 60% reductions in anthropogenic NO_x and VOC emissions would be required to reach the 2008 ozone air quality standard (i.e., 75 parts per billion (ppb)) for several U.S. urban areas in 2006.

Satellite-retrieved formaldehyde (HCHO) and nitrogen dioxide (NO_2) column densities have been widely applied to investigate spatial and temporal changes in VOC and NO_x emission rates, respectively (Russell et al., 2012; Streets et al., 2013; Zhu et al., 2014). HCHO mainly comes from oxidation of non-Methane volatile organic compounds (NMVOCs) which have a short lifetime in the atmosphere. HCHO can reflect VOC emissions over vegetated and biomass burning areas through fast production from short-lived NMVOCs, but it is limited to reflect anthropogenic VOC emissions except in highly-polluted urban areas (Streets et al., 2013). The ratio of HCHO to NO_2 column densities (i.e., [HCHO]/[NO_2]) can indicate the NO_x- or VOC-limited ambient ozone formation regime over a long time horizon (i.e., 5–10 years) when their corresponding [HCHO]/[NO_2] are determined using photochemical models (Choi et al., 2012; Duncan et al., 2010; Martin et al., 2004). [HCHO]/[NO_2] is an effective function to reflect surface ozone chemistry using retrievals from satellite instruments, since HCHO and NO_2 columns are detected within the mixed layer (Ladstätter-Weißenmayer et al., 2003) and is also strongly related to the reactivity-weighted VOC/NO_x ratios (Chameides et al., 1992). In a study by Martin et al. (2004), [HCHO]/[NO_2] for the NO_x-limited and VOC-limited ozone formation regimes were determined using the Global Ozone Monitoring Experiment (GOME)-retrieved column densities and the GEOS-Chem model. [HCHO]/[NO_2] for three different chemical conditions (i.e., NO_x-limited, VOC-limited and transition regimes) were also determined by Duncan et al. (2010) using Ozone Monitoring Instrument (OMI)-retrieved NO_2 columns and a photochemical box model. Choi et al. (2012) conducted a similar study but using Global Ozone Monitoring Experiment-2 (GOME-2) column densities and regional air quality modeling. Choi and Souri (2015) also examined OMI-retrieved NO_2 and HCHO column densities, and the results show ambient ozone formation in Texas urban areas became more NO_x-sensitive from 2005 to 2013 due to controls of anthropogenic NO_x emissions. In this study, a state-of-the-art photochemical model, the Community Multiscale Air Quality (CMAQ) (Byun and Schere, 2006) and retrievals from two satellite instruments were used together to determine [HCHO]/[NO_2] for VOC- and NO_x-limited ambient ozone formation regimes. This work leveraged: (1) satellite-retrieved HCHO and NO_2 column densities from multiple instruments; (2) photochemical air quality modeling and sensitivity analysis; and (3) ratios of satellite-retrieved air pollutant column densities to investigate long-term variations in ambient ozone formation regimes for neighboring cities of shale plays in the Northeast United States (U.S.) (Fig. 1(b)). Satellite-retrieved HCHO and NO_2 column densities from a single instrument were used in previous studies (Choi et al., 2012; Duncan et al., 2010; Martin et al., 2004). In this study, we used HCHO and NO_2 column densities measured by OMI and GOME-2 to examine differences in satellite-measured air quality between two different satellite instruments. The purpose is to understand the variations of retrieved products from operating limits of the instruments in order to avoid the bias of using a single instrument. To the best of our knowledge, this is the first study to focus on the difference between satellite retrieved HCHO and NO_2 column densities and to diagnose

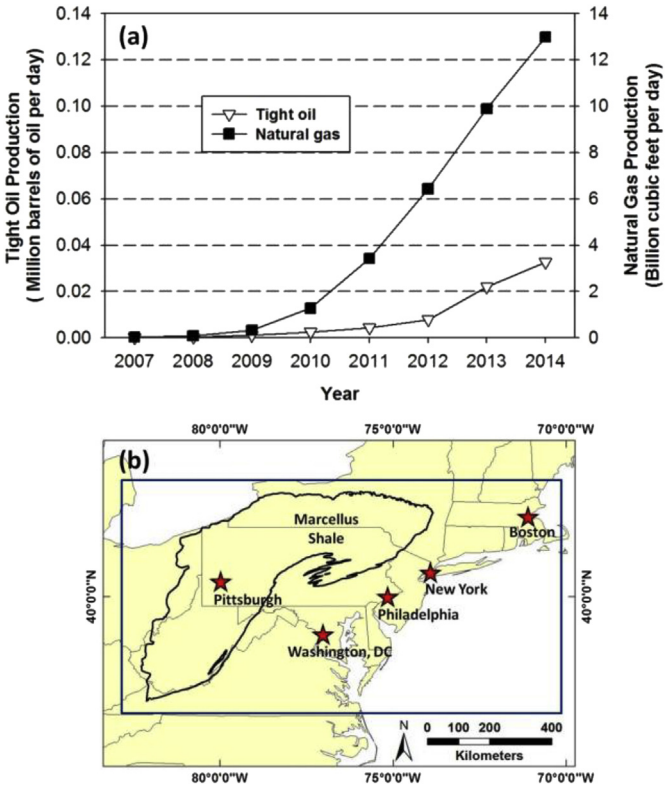

Fig. 1. (a) Tight oil and natural gas production from Marcellus Shale. (b) Locations of neighboring cities (i.e., Boston, New York, Philadelphia, Pittsburgh and Washington, DC) of Marcellus Shale in the Northeast U.S.

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