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Staggering reductions in atmospheric nitrogen dioxide across Canada in response to legislated transportation emissions reductions

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HIGHLIGHTS

- Ambient NO_2 levels in Canada currently vary between 1.16 and 14.96 ppb, with the national average being 8.43 ppb.
- Mann-Kendall long-term trend analysis indicates that NO_2 levels have significantly decreased at 87% of sites.
- Declining transportation emissions have had the largest influence on decreasing NO_2 levels.
- Changepoint detection shows that legislated transportation emissions reductions lead to subsequent decreases in NO_2 levels.
- There is ~2 years of lag between emissions reductions and the reporting of reductions.

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ABSTRACT

It is well established that atmospheric nitrogen dioxide (NO_2), associated mainly with emissions from transportation and industry, can have adverse effects on both human and ecosystem health. Specifically, atmospheric NO_2 plays a role in the formation of ozone, and in acidic and nutrient deposition. As such, international agreements and national legislation, such as the *On-Road Vehicle and Engine Emission Regulations (SOR/2003-2)*, and the *Federal Agenda on Cleaner Vehicles, Engines and Fuel* have been put into place to regulate and limit oxidized nitrogen emissions. The objective of this study was to assess the response of ambient air concentrations of NO_2 across Canada to emissions regulations. Current NO_2 levels across Canada were examined at 137 monitoring sites, and long-term annual and quarterly trends were evaluated for 63 continuous monitoring stations that had at least 10 years of data during the period 1988–2013. A non-parametric Mann-Kendall test (Z values) and Sen's slope estimate were used to determine monotonic trends; further changepoint analysis was used to determine periods with significant changes in NO_2 air concentration and emissions time-series data. Current annual average NO_2 levels in Canada range between 1.16 and 14.96 ppb, with the national average being 8.43 ppb. Provincially, average NO_2 ranges between 3.77 and 9.25 ppb, with Ontario and British Columbia having the highest ambient levels of NO_2. Long-term tend analysis indicated that the annual average NO_2 air concentration decreased significantly at 87% of the stations (55 of 63), and decreased non-significantly at 10% (5 of 63) during the period 1998–2013. Concentrations increased (non-significantly) at only 3% (2 of 63) of the sites. Quarterly long-term trends showed similar results; significant decreases occurred at 84% (January–March), 88% (April–June), 83% (July–September), and 81% (October–December) of the sites. Declines in transportation emissions had the most influence on NO_2 air concentrations, and changepoint analysis identified three significant changepoints for the air concentration of NO_2 and transportation emissions data. The air concentration changepoints occurred immediately following changepoints in transportation emissions. The introduction of emissions limiting legislation, primarily from transportation sources, has lead to dramatic decreases of 32% in NO_x emissions (42% from transportation sources [road, rail, air, marine]) and 47% in ambient NO_2 concentrations across Canada. With respect to human health, legislated changes in transportation emissions have the greatest impact on ambient concentration in urban areas.
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1. Introduction

Atmospheric nitrogen dioxide (NO_2) is associated mainly with

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emissions from transportation and industrial sources, specifically from combustion-engines. Ambient NO_2 concentrations are primarily derived from secondary oxidation reactions of nitrogen oxides (NO_x) in the atmosphere (Mavroidis and Chaloulakou, 2011). Nitrogen dioxide is a strong oxidizing agent, reacting with hydroxyl radical (OH) in the atmospheric to form nitric acid (HNO_3), which can directly contribute to acidification (Bouwman et al., 2002). Nitric acid is also a component of smog, and can directly impact human respiratory health (Latza et al., 2009).

Canadian air monitoring networks (e.g., National Air Pollution Surveillance (NAPS) Program) have been established to monitor and assess ambient atmospheric pollutants, including NO_2. These networks are used to determine spatial and temporal patterns, to monitor atmospheric levels and warn of potential health or environmental impacts, and to help determine policy changes. Increasing awareness of the health and environmental impacts associated with nitrogen (N) emissions have lead to national, and international, regulations and agreements to control emissions. Agreements such as the Canada-U.S. Air Quality Agreement, and the Clean Air Act, limit the amount of N that is being emitted and transported across international boundaries. Similarly, regulations such as the Canadian *On-Road Vehicle and Engine Emission Regulations (SOR/2003-2)*, directly control NO_x emission from transportation and industry.

It is important to evaluate how pollutant levels have responded to policy change to determine the efficacy of these legislative decisions. However few studies have assessed long-term air quality in Canada or trends for NO_2, as monitoring data is primarily used to report current pollutant levels, and to deliver real-time air quality and health information. The objective of this study was to assess the long-term response of ambient air concentrations of tropospheric NO_2 across Canada to emissions regulations.

The non-parametric Mann-Kendall test was used to determine monotonic trends in the annual average concentration of NO_2 across Canada during the period 1988–2013. In addition, change-point detection was preformed to determine significant changes in time-series NO_2 air concentration and emissions data. We specifically ask two questions (1) how quickly do air concentrations respond to legislated emissions reductions, and (2) what emissions sources are the principal driver of atmospheric change?

2. Methods

2.1. Study sites

The NAPS Program has 286 stations (as of 2014) located in 203 communities across Canada. The network covers all provinces and territories, and stations are selected to be representative of the Canadian population (Environment Canada, 2014). NAPS stations are primarily used to monitor air quality and to provide an air quality health index measurement; as a result sites are mainly located in populous, urban areas. In addition, a large proportion of NAPS sites are located in Southern Ontario and Quebec (Fig. 1), along the country's largest and busiest highway. There are 137 NAPS stations with atmospheric NO_2 data available during the period 2009–2013. In this study, these sites were selected to represent current NO_2 levels across Canada (current sites). Further, stations that had ≥90% data completeness for a minimum of 10 years, between 1988 and 2013, were selected for long-term trend analysis (long-term sites). Data series from several monitoring stations were combined into one unified data time-series to accommodate data gaps. These stations were geographically close together, and had overlapping data windows within the same year; this indicated that one site had opened, and another closed, during a calendar year. This yielded 63 long-term sites for trend analysis across Canada (Fig. 1).

2.2. Data sources

Data were obtained from the NAPS Program (www.ec.gc.ca/rnspa–naps). The Program monitors ambient atmospheric NO_2 in real-time, with concentrations in parts per billion (ppb) recorded every hour using chemiluminescence analyzers (Environment Canada, 2014). Nitric oxide (NO) and NO_x ($NO + NO_2$) are measured together at each site, then NO_x is reduced and NO_2 levels are calculated using subtraction. All available data during the period 1988–2013 for all the stations in Canada was downloaded for examination in this study.

The data were examined to determine eligibility for current NO_2 levels in Canada. Only stations that had at least one annual data value during 2012–2013 were retained (n = 137). The data were further examined to determine the data window for long-term trend analysis. The criterion for inclusion was at least 10 years of monitoring data during the period 1988–2013. The data were also examined for completeness; only years that had ≥90% completeness were retained for long-term trend analysis. Quarterly data were processed in a similar manner. After checking for data completeness, each station's hourly data was split into three-month periods (representing quarters), for each year of the data time-series. Quarters were determined as: Q1 = January–March, Q2 = April–June, Q3 = July–September and Q4 = October–December.

Provincial and national population data were obtained from Statistics Canada (www.statcan.gc.ca). Nitrogen oxide emissions data were obtained from Environment Canada (www.ec.gc.ca/indicateurs-indicators), and land-use type for each monitoring station was obtained from the NAPS data (included in the metadata for the sites).

2.3. Data analysis

The current annual average levels of ambient NO_2 nationwide were determined for the period 2009–2013 (five-year annual average) for each station (137 current sites). The average, median, and 5th and 95th percentile NO_2 concentration was calculated for each province/territory. The coefficient of variation (COV), i.e., standard deviation divided by the mean and multiplied by 100, was also calculated.

The annual average concentration at each of the long-term sites was calculated; hourly NO_2 concentration data for the entire year were averaged into one annual average value per year, per site, for the period 1988–2013. Similarly, quarterly average values were calculated; the hourly NO_2 concentration values were averaged for each site, for each quarter, per year.

The Mann-Kendall non-parametric test (Mann, 1945) was used to assess monotonic trends in the average annual air concentration of NO_2 at the 63 long-term NAPS monitoring sites, during the period 1988–2013. The Mann-Kendall test is a non-parametric test used to determine if time series data exhibits an increasing, or decreasing trend. It is not dependent on assumptions of distribution, missing data, or irregularly spaced monitoring periods, and as such is useful for long term atmospheric pollutant monitoring data, which is often non-normal, missing data, and has inconsistent periods of data collection. The Mann-Kendall statistic has been used widely to assess long-term trends in atmospheric concentrations (Chaudhuri and Dutta, 2014; Hole et al., 2009; Saylor et al.,

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