

Examination of temporal and spatial variability of NO₂ VCDs measured using mobile-MAX-DOAS in Toronto, Canada.

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Mobile-MAX-DOAS is an innovative technique used to estimate pollutant emission rates and validate satellite measurements and air quality models. It is essential to identify and examine factors that can significantly impact the accuracy of this developing technique.

Mobile-MAX-DOAS measurements were conducted in Toronto, Canada with a mini-MAX-DOAS instrument mounted (pointing backwards) on top of a car during August and September, 2016. Scattered sunlight spectra were collected every 45 seconds in the continuously repeated sequence of elevation angles of 30°, 30°, 30°, 30°, 40°, 30°, 90°. Tropospheric VCDs were determined using the geometric approximation from DSCDs fitted using a near-noon, low NO₂ VCD FRS spectrum. The study goal was to examine the validity of the assumption that VCDs remain relatively constant at each measured location on a driving route encircling an urban area of interest with typical time periods of 1.5-3 hours to estimate emissions and whether driving direction significantly impacts results. NO₂ VCD temporal variability was therefore determined by repeating driving routes in both directions in quick succession on multiple days.

Strong temporal variability in NO₂ VCDs of up to a factor of two were observed for some routes for the same vehicle locations under constant prevailing wind conditions within <2 hours. These differences may be due to the effects of transport, changing tropospheric chemistry and/or diurnal trends in emissions rates. Under these conditions measurements along different portions of the encircled area in a large city may not be representative of the entire measurement period, introducing error into the final emission estimate.

Certain straight roads exhibited significantly different VCDs within < 30 minutes when the instrument azimuth pointing direction was changed by 180°. The weighted average VCD was $\sim 8(\pm 3) \times 10^{16}$ molec. cm⁻¹ from driving in one direction but $\sim 4(\pm 1.5) \times 10^{16}$ molec. cm⁻¹ from driving in the opposite direction. This indicates sufficient horizontal inhomogeneity for the instrument to view significantly different NO₂ regimes while at the same vehicle geographical location due to the different azimuth direction.

NO₂ line fluxes were determined during weekday afternoon rush-hours by driving repeatedly in both directions under tangential prevailing winds conditions on a road that is 8km downwind of Toronto and 4km downwind of a major highway. During one afternoon the average NO₂ VCD was $6(\pm 2) \times 10^{16}$ molec. cm⁻² with a standard deviation of 3×10^{15} molec. cm⁻². This average value is consistent with NO₂ VCDs retrieved using optimal estimation methods from stationary MAX-DOAS measurements at nearby York University. Using a 10m elevation measured wind-speed of 16km hr⁻¹, the NO₂ line flux was $3(\pm 9) \times 10^{18}$ molec. cm⁻¹s⁻¹, approximately 6 times that determined by Halla et al. (2011) for a line flux measured in a rural area of southwestern Ontario, downwind of pollution sources in Michigan and Ohio. Our resulting average NO₂ flux of 84 (+/-25) mg m⁻²hr⁻¹ is consistent with NO_x fluxes measured in major European cities of up to 90 mg m⁻²hr⁻¹.

This work will be used as a baseline experiment to apply this method in other Canadian cities.