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Ten-year Trends in Ambient $PM_{2.5}$ and $PM_{2.5}$-bound Element Concentrations in an Industrialized Border City

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Abstract **-** In this study, linear regression was adopted to characterize trends in ambient fine particulate matter ($PM_{2.5}$) and $PM_{2.5}$-bounded element concentrations using data collected at Windsor West monitoring station in Windsor, Ontario, Canada, during 2003-2012. Twenty-two $PM_{2.5}$-buonded elements were reported, and 10 of them had concentrations below method detection limits over 70% of the time. Therefore, they were excluded from further analysis. The 12 retained elements are Al, Br, Ca, Fe, K, Mn, Ni, Pb, S, Si, Ti and Zn. These 12 elements combined contributed 13% of total $PM_{2.5}$ concentration, with S being the largest contributor (8.4%), followed by Fe (1.4%). The annual mean $PM_{2.5}$ concentrations ranged from 7.3 μg/m^3 in 2009 to 10.4 μg/m^3 in 2005, are lower than the Canadian Ambient Air Quality Standards of 10 μg/m^3 for $PM_{2.5}$ except for in 2005. The ten-year mean was 8.8 μg/m^3. Significant decreasing trends were found for $PM_{2.5}$ (25%), Fe (19%), Mn (47%), S (44%), Si (72%), Ti (92%), and Zn (71%), while no statistically significant change was observed for Al, Br, Ca, K, Ni, and Pb. Our findings suggest that the emission control strategies implemented during the study period were effective in reducing concentrations of $PM_{2.5}$ and six out of 12 $PM_{2.5}$-bounded elements in Windsor.

Keywords**:** Fine particulate matter ($PM_{2.5}$), $PM_{2.5}$-bound elements, Long-term trend, Windsor

1. Introduction

$PM_{2.5}$, also known as fine inhalable particles, refers to fine particulate matter with aerodynamic diameters that are 2.5 micrometers and smaller. Major sources of $PM_{2.5}$ include coal-fired power plants, industrial processes, transportation, biomass burning, and dust. Exposure to ambient $PM_{2.5}$ is associated with various human health effects such as respiratory and cardiovascular diseases, lung cancer, and premature death [1]. High $PM_{2.5}$ concentrations can cause impaired visibility, often observed in urban centers of some developing countries.

Winsor is an industrial city in southwestern Ontario, Canada directly across from Detroit, Michigan, U.S. $PM_{2.5}$ concentrations in Windsor are impacted by local emissions and transboundary input [2]. From 2003 to 2012, the annual $PM_{2.5}$ concentrations decreased 31% from 2003-2012 [2]. However, concentrations of $PM_{2.5}$-bound elements are better indicators of health effects due to their toxicity. For example, exposure to nickel could lead to lung and kidney damage [3]. This study investigates the 10-year trends of $PM_{2.5}$ and $PM_{2.5}$-bound element concentrations in Windsor during 2003-2012.

2. Methodology

2.1. Data Sources

Concentrations of ambient $PM_{2.5}$ and $PM_{2.5}$-bound elements were monitored at Windsor West station (Fig. 1) by Ontario Ministry of the Environment, Conservation and Parks under the National Air Pollution Surveillance program [4]. The station is classified as urban site type and it is surrounded by green space and two-story residential buildings. The station is in close proximity to the Huron Church Road and the Ambassador Bridge, the busiest international border crossing in the North America [5].

Continuous $PM_{2.5}$ concentrations was measured using Tapered Element Oscillating Microbalance equipped with sample equilibration system (Thermo Fisher Scientific Inc.) The monthly concentration data during the study period of 2003-2012 were download from ECCC (Environment and Climate Change Canada) website [4]. For $PM_{2.5}$-bound elements, air filters were collected with dichotomous (Dichot) samplers. The 24-hour intergraded samples were collected once every 6-day. Microbalance was employed to weigh the total $PM_{2.5}$ mass collected on each filter. XRF (X-ray fluorescence) was used to detect 22 elements, Al, Ba, Br, Ca, Cd, Cr, Cs, Fe, K, Mn, Ni, Pb, Rb, S, Sb, Se, Si, Sn, Sr, Ti, V, and Zn. Concentrations of $PM_{2.5}$-bound elements were download from ECCC website [4].

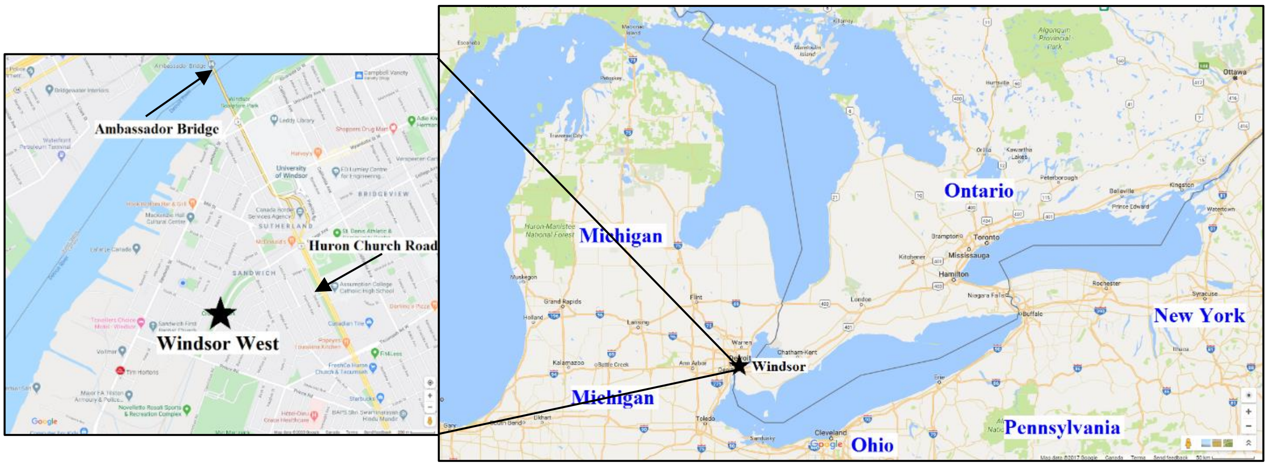

Fig. 1: Location of the Windsor West air monitoring station in Ontario, Canada.

2.2. Data Analysis

$PM_{2.5}$ and $PM_{2.5}$-bound element concentrations were screened by counting the percentages of missing, below method detection limits, and flagged data points. Ten out of 22 elements were excluded from further analysis due to more than 70% samples were below method detection limit. The remaining 12 elements are Al, Br, Ca, Fe, K, Mn, Ni, Pb, S, Si, Ti, and Zn. Percentage contributions of each element to $PM_{2.5}$ mass were calculated. Annual means of monthly $PM_{2.5}$ and 24-hour $PM_{2.5}$-bound element concentrations were used to assess long-term trend with linear regression.

3. Results and Discussion

From 2003 to 2012, the annual $PM_{2.5}$ concentrations were less than the Canadian Ambient Air Quality Standards (CAAQS) of 10 μg/m³ [6] except for 2005 when the concentration was 10.4 μg/m³ (Fig. 2). The 10-year average was 8.8 μg/m³. The annual concentrations decreased from 9.6 μg/m³ in 2003 to 7.7 μg/m³ in 2012.

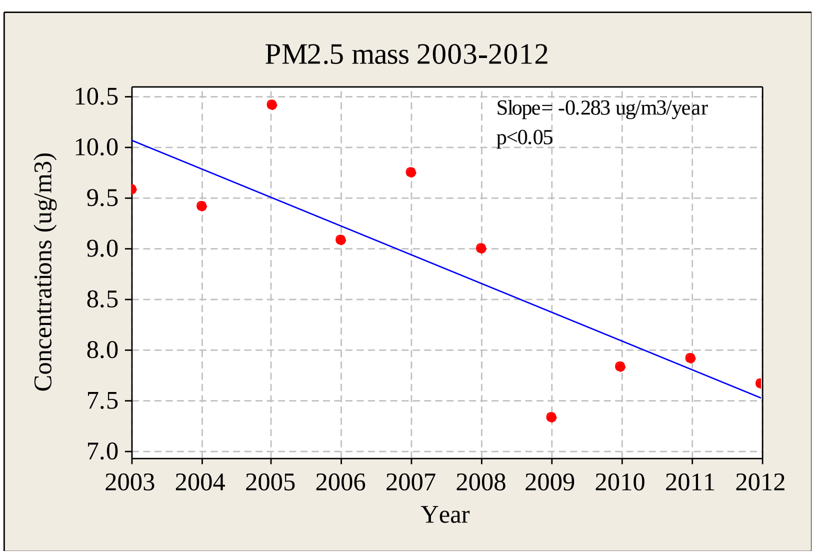

Fig. 2: Annual $PM_{2.5}$ concentrations.

As shown in Table 1, concentrations of $PM_{2.5}$, Fe, Mn, Si, Zn decreased significantly ($p<0.05$) during the 10-year study period, by 25, 19, 47, 72, and 71%, respectively. The rates of per-year change were -0.28 µg/m^3, -5.5 ng/m^3, -1.1 ng/m^3, -9.1 ng/m^3, and -3.2 ng/m^3, respectively. There were marginally ($p<0.1$) decreases in concentrations of S (-69 ng/m^3/year) and Ti (-1.6 ng/m^3/year). Al, Br, Ca, K, and Ni experienced decreased concentrations. However, these trends were not statistically significant ($p>0.1$). Pb is the only element showed no trend.

Table 1: Ten-year mean concentrations and trends of $PM_{2.5}$ and 12 $PM_{2.5}$-bound elements in Windsor. Changes in percentage were estimated using the slopes.

$PM_{2.5}$	**10-year mean (µg/m^3)**	**Slope (µg/m^3/year)**	**Change in 10-year (%)**	**p-value**
$PM_{2.5}$	8.8	-0.28	-25	<0.05
Elements	**10-year mean (ng/m^3)**	**Slope (ng/m^3/year)**	**Change in 10-year (%)**	**p-value**
Fe	127	-5.5	-19	<0.05
Mn	7.86	-1.1	-47	<0.05
Si	75.5	-9.1	-72	<0.05
Zn	28.0	-3.2	-71	<0.05
S	997	-69	-44	<0.1
Ti	7.08	-1.6	-92	<0.1
Al	35.1	-1.2	-31	>0.1
Br	2.90	-0.037	-8.6	>0.1
Ca	62.7	-2.7	-33	>0.1
K	69.1	-1.6	-16	>0.1
Ni	2.51	-0.22	-47	>0.1
Pb	5.73	0.042	9.2	>0.1

Among the 12 elements considered, S is the largest contributor to $PM_{2.5}$ (10-year average 8.4%, Fig. 3). This is in agreement with findings of other studies, e.g., 10% in Calgary during 2015-2019 [7]. The second largest contributor was Fe (1.4%). K, Si, Ca, Al, Zn contributed less than 1% each, while Pb, Mn, Ti, Ni, Br contributed less than 0.1% each. When combined, these 12 elements contributed 13% to total $PM_{2.5}$.

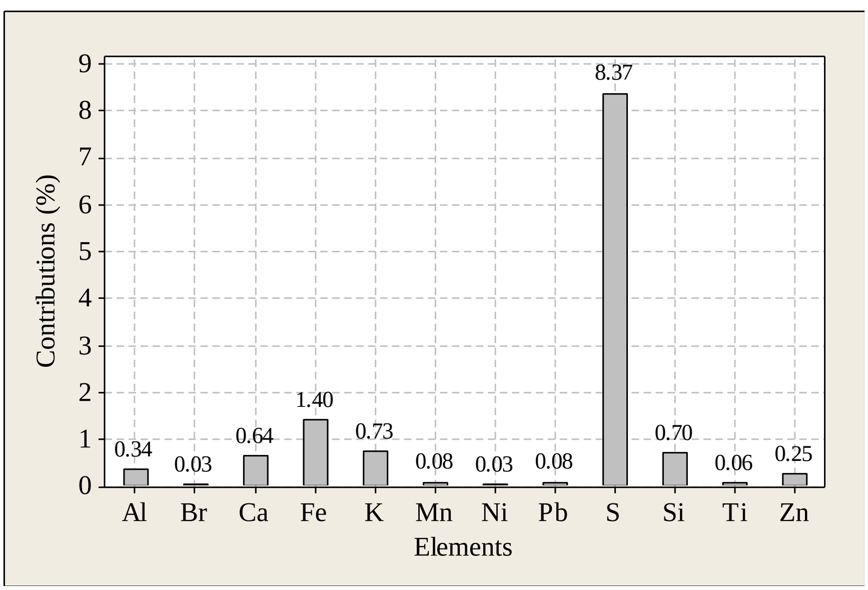

Fig. 3: Element contributions to total $PM_{2.5}$ concentrations.

There were year-to-year variations in individual element contributions. From 2003 to 2012, Ni and Si has decreased contributions, while Br was the only element showed increased contributions. Contributions from Al and Mn decreased 2008 then increased and remained stable, respectively. No trend was apparent for Ca, Fe, K, Pb, S, Ti, and Zn.

4. Conclusions

In this study, ambient concentrations of $PM_{2.5}$ and $PM_{2.5}$-bound elements collected in Windsor from 2003 to 2012 were analysed to assess long-term trend. The 10-year average $PM_{2.5}$ concentrations was 8.8 µg/m^3, below the CAAQS of 10 µg/m^3. Nine out of 10 annual means were also below the CAAQS. The annual $PM_{2.5}$ concentrations decreased significantly by 0.28 µg/m^3 per year. Concentrations of 11 out of 12 elements also decreased, with rates ranging from -0.04 to -69 ng/m^3/year, while the changes were statistically insignificant for Al, Br, Ca, K, and Ni. No clear trend was observed for Pb. The ranking of element contribution to total $PM_{2.5}$ was S (8.4%) > Fe (1.4%) > K, Si, Ca, Al, Zn (<1% each) > Pb, Mn, Ti, Ni, Br (<0.1% each). The total contributions from the 12 elements averaged 13%. Results of our study indicate that emission control implemented during the study period was effective in reducing concentrations of $PM_{2.5}$ and six out of 12 elements. However, the effect is limited for Pb. Further efforts are needed in reducing Pb and S emissions in Windsor and surrounding areas in Canada and the US.

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