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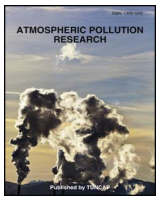

Characterization of air quality and sources of fine particulate matter ($PM_{2.5}$) in the City of Calgary, Canada

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ABSTRACT

With concern about fine particulate matter ($PM_{2.5}$) pollution in urban areas and levels approaching a new Canadian Ambient Air Quality Standards (CAAQS), an exploratory study of air quality characteristics and potential sources affecting $PM_{2.5}$ levels was undertaken in the City of Calgary, Alberta. The study was performed for the economic recession period 2014 to 2016 using hourly concentrations of criteria air pollutants at two monitoring stations (Calgary central and Calgary northwest). The overall mean and median $PM_{2.5}$ concentrations were similar at both Calgary central (arithmetic mean: 7.7 μg/m^3, median: 6.0 μg/m^3) and Calgary northwest (arithmetic mean: 7.5 μg/m^3, median: 6.0 μg/m^3). Three-year averages of annual average daily 24 h $PM_{2.5}$ concentrations at both stations were below the 2015 annual CAAQS of 10 μg/m^3 during the study period 2014–2016. A multivariate receptor model positive matrix factorization (PMF) revealed five sources, where secondary aerosol was identified as the largest source of $PM_{2.5}$ contributing 54% at Calgary central and 42% at Calgary northwest. Other sources included combustion (18%, 39%), traffic (18%, 12%), an O_3-rich source (8%, 4%), and a mixed urban source (2%, 3%) at Calgary central and Calgary northwest, respectively. Variations in annual contributions of secondary aerosol, combustion and traffic were observed at Calgary downtown for 2014–2016. At Calgary northwest no variation was found for annual traffic contributions. These findings offer preliminary information about the contributions of different potential sources to $PM_{2.5}$ in Calgary; and this information can support policy makers in developing appropriate air quality management initiatives for $PM_{2.5}$ pollution if needed.

1. Introduction

There has been growing awareness and public health concerns about the state of air quality in urban areas. Urban air pollution is generally caused by a wide variety of emission sources including traffic, industry, commercial/residential fuel combustion and is comprised of a complex mixture of gaseous and particulate air contaminants such as nitrogen dioxide (NO_2), sulfur dioxide (SO_2), fine particulate matter ($PM_{2.5}$), and ground-level ozone (O_3). Epidemiological studies suggest potential associations between short- and long-term exposure to criteria air pollutants and increased morbidity, mortality and hospital admissions for cardiovascular and pulmonary diseases, stroke, as well as decreased life expectancy (Burnett et al., 1999, 2004; Ruidavets et al., 2005; Pope et al., 2014; Weichenthal et al., 2014; Zanobetti et al., 2014). High concentrations of these pollutants can also contribute to acid deposition, photochemical smog and reduced atmospheric visibility (Cooper and Alley, 2002; Cheung et al., 2005). This has led the Canadian Council of Ministers of the Environment (CCME) to the establishment of health-based air quality standards i.e., Canadian Ambient Air Quality Standards (CAAQS) as a driver for air quality management across the country with an objective to guide work towards better understanding and, where necessary, controlling air emissions in populated urban areas. The new CAAQS for $PM_{2.5}$ for the year 2015 (annual: 10 μg/m^3, 24 h: 28 μg/m^3) and 2020 (annual: 8.8 μg/m^3, 24 h: 27 μg/m^3) (CCME, 2012) replaced the former 24 h Canada-Wide Standard (CWS) of 30 μg/m^3 established in 2000 (CCME, 2000).

The City of Calgary is the largest urban area and most populous city (area 825 km^2, population 1,246,337, Municipal census, 2017) in the oil and natural gas-rich province of Alberta and third-largest municipality in Canada (Statistics Canada, 2016). It is located about 300 km south of Alberta's capital Edmonton in a valley at the foothills approximately 80 km east of the front ranges of the Canadian Rockies. The city anchors the south end of the Calgary-Edmonton Corridor, which is home to 2.7 million people. Alberta has well-established conventional oil and gas extraction, refining and upgrading activities in addition to unconventional oil sands development in the northeast area of the

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province. Calgary is located within the southern edge of oil and gas extraction activities (Supplemental Information-SI, Fig. S1a). Because of its diversified economy including oil and gas, film and television industries, transportation and logistics, technology, manufacturing, retail, and tourism sectors, Calgary plays a key role in supporting economic growth of Alberta and Canada.

Evaluation of long-term air monitoring data and characterization of ambient $PM_{2.5}$ can aid in improving the understanding of the state of air quality and sources of particle pollution in urban areas. In our recent study (Bari and Kindzierski, 2016a), we observed more than 40 (80) exceedances of the 24 h $PM_{2.5}$ CWS of 30 μg/m^3 (1 h Alberta Ambient Air Quality Guideline of 80 μg/m^3), respectively over a 17-year period (1998–2014) in Calgary. In addition, the highest 3-year average (2010–2012) of 24 h concentrations was recorded in downtown Calgary among Canadian urban areas. In another study conducted for the time period 2010–2012 in six Alberta airsheds including South Saskatchewan Air Zone where Calgary is located (Fig. S1b), the Government of Alberta (Alberta Environment and Parks-AEP, 2015) reported that air monitoring stations at Calgary central and Calgary northwest had annual $PM_{2.5}$ metric values of 7.5 μg/m^3 (in 2013) and 8.5 μg/m^3 (3-year average). The study assigned Calgary to an orange management level for $PM_{2.5}$ based on four-color coded air quality management thresholds for 2015 (Table S1), suggesting that $PM_{2.5}$ concentrations were approaching the new CAAQS and proactive planning and/or action may be needed to prevent exceedances. It was therefore of interest to undertake an exploratory study to evaluate $PM_{2.5}$ levels and to identify different emission sources that affect $PM_{2.5}$ levels in Calgary.

In general, 24 h $PM_{2.5}$ chemical speciation data has been widely used in multivariate receptor models to identify and distinguish different emission sources in urban areas. Due to cost, however many monitoring organizations do not have the capabilities and financial resources to routinely monitor for $PM_{2.5}$ chemical species. Environment and Climate Change Canada (ECCC) only performs $PM_{2.5}$ speciation monitoring in selected major urban centers of Canada (e.g., Edmonton, Toronto, Vancouver, Montreal) (ECCC, 2017a; Bari and Kindzierski, 2016b). In Alberta local airshed monitoring organizations work collaboratively with Alberta Environment and Parks to operate air quality monitoring networks and monitoring stations in cities, small towns and rural areas and measure real-time concentrations of gaseous pollutants and $PM_{2.5}$. Due to lack of $PM_{2.5}$ speciation data, numerous studies have been carried out worldwide to characterize sources of ambient fine particulate matter, nanoparticles, particle size distribution using real-time concentrations of gaseous pollutants data (e.g., Yue et al., 2008; Thimmaiah et al., 2009; Hellebust et al., 2010; Sun et al., 2014; Khan et al., 2015; Al-Dabbous and Kumar, 2015; Sowlat et al., 2016). Using only real-time gaseous pollutant data in receptor models may provide a limited number of source factors and may not be able to identify some specific sources (e.g., road dust, secondary organic aerosol, biogenic). In addition, this approach may also not be able to distinguish different industry-related sources (e.g., metallurgy, refinery, cement kiln) that can be important sources in urban areas of Alberta. However, the approach can offer useful preliminary information highlighting potential major emission source types that affect air quality at a receptor location in urban areas. In our recent studies (Bari and Kindzierski, 2017a,b), we used the positive matrix factorization (PMF) model to investigate $PM_{2.5}$ sources in the third largest urban area (Red Deer) and a small rural community (Hinton) in Alberta using gaseous pollutant data. In this exploratory study, we characterized air quality and investigated emission sources that affect $PM_{2.5}$ levels in the largest city of Alberta – Calgary using real-time continuous air monitoring data.

2. Methodology

2.1. Study areas

As part of ECCC's National Air Pollution Surveillance (NAPS) initiatives, the Calgary Region Airshed Zone Society (CRAZ, http://www.craz.ca) has been responsible for regional air quality monitoring and providing results to Alberta Environment and Parks. The CRAZ airshed boundaries include the cities of Calgary and Airdrie, the Municipal Districts of Rocky View, Bighorn and Foothills, Willow Creek, Vulcan and Wheatland Counties, the Improvement Districts of Kananaskis and Banff, and the Town of Canmore (Fig. S1c). The topographical map of the City of Calgary and immediate surrounding area is shown in Fig. S2a. Average daily (24 h) weekday traffic volume for the City of Calgary in 2016 is shown in Fig. S2b.

Calgary has a humid continental climate with wide fluctuations in temperatures throughout the seasons e.g., long cold winters and warm summers and no dry season. Seasonal average daily temperatures typically range from −6.8 °C in December to 16.5 °C in July and the average annual precipitation is in terms of rain 33 cm and snow 129 cm (ECCC, 2017b). Due to close proximity to the Rocky Mountains, Calgary's air quality can be influenced by warm and dry Chinook winds that blow over the mountains during winter months (Hicks and Mathews, 1979). Winter temperatures are also affected by the wind chill factor, with a high average wind speed of 14.2 km/h in Calgary, one of the highest in Canadian cities. Early morning ground-based temperature inversions are common throughout the whole year in Alberta with deeper and stronger inversions observed during winter months (Hicks et al., 1977; Myrick et al., 1994), thus limiting the dispersion of air pollutants and potentially increasing pollutants levels in winter.

According to Environment Canada 2006 and CRAZ 2008 emissions inventories, major point sources of air pollutant emissions in the CRAZ region are several upstream oil and gas facilities, cement and other industrial activities (e.g., construction). There are also non-point sources e.g., transportation, residential and commercial heating and open area sources including agricultural operations and field/stock burning as well as solvent and biogenic emissions (Novus Environmental, 2013). In the City of Calgary, contributions to total emissions were noteworthy for transportation (carbon monoxide, CO: 89% of total emissions, oxides of nitrogen, NO_X: 79%, $PM_{2.5}$: 9%), construction related fugitive emissions ($PM_{2.5}$: 62%), cement industry (sulfur dioxide, SO_2: 40%) and commercial and residential heating (SO_2: 28%, $PM_{2.5}$: 9%) (Table S2). National Pollutant Release Inventory (NPRI) (ECCC, 2017c) reported annual releases of SO_2, NO_X, $PM_{2.5}$, and volatile organic compounds (VOCs) to the air from major industrial facilities within 50 km of downtown Calgary are shown in Table S3. Notable energy development activities are upstream oil and gas developments located within and surrounding the City of Calgary. For example, several oil and gas industries such as Inter Pipeline Extraction Ltd.-Cochrane Extraction Plant, Taqa North Ltd.-East Crossfield Gas Plant, Enmax Generation Portfolio Inc.-Shepard Energy Centre and Direct Energy Marketing Ltd.-Wildcat Hills Gas Plant are located from 35 km northwest, 39 km north, 16 km southeast and 44 km northwest from Calgary central, respectively (Table S3). Over the 10-year period 2004–2014 the city added 261,700 residents to its population (City of Calgary, 2015), and 26,170 more registered vehicles each year (average) using its roadways (Alberta Transportation, 2015).

The study was performed using historical air quality data collected from two air monitoring stations – one in downtown Calgary i.e., Calgary central (51.0472° N, 114.0731° W, elevation 1051 m) and one in a residential area in Calgary northwest (51.0792° N, 114.1419° W, elevation 1106 m) (Fig. 1). The local emission sources at Calgary central are traffic and commercial operations, while at Calgary northwest potential sources include residential heating, traffic and small-scale industries (e.g., cement industry located within 10 km northwest from the Calgary northwest station, Table S3). Other industries are located to the east and southeast of Calgary within 5–10 km from Calgary downtown. Over the last 10 years, Calgary downtown station was moved ~100 m west to Calgary central 2 (51.0459°N, 114.0747° W) on April 2008 and 4.9 km southeast to Calgary-central Inglewood

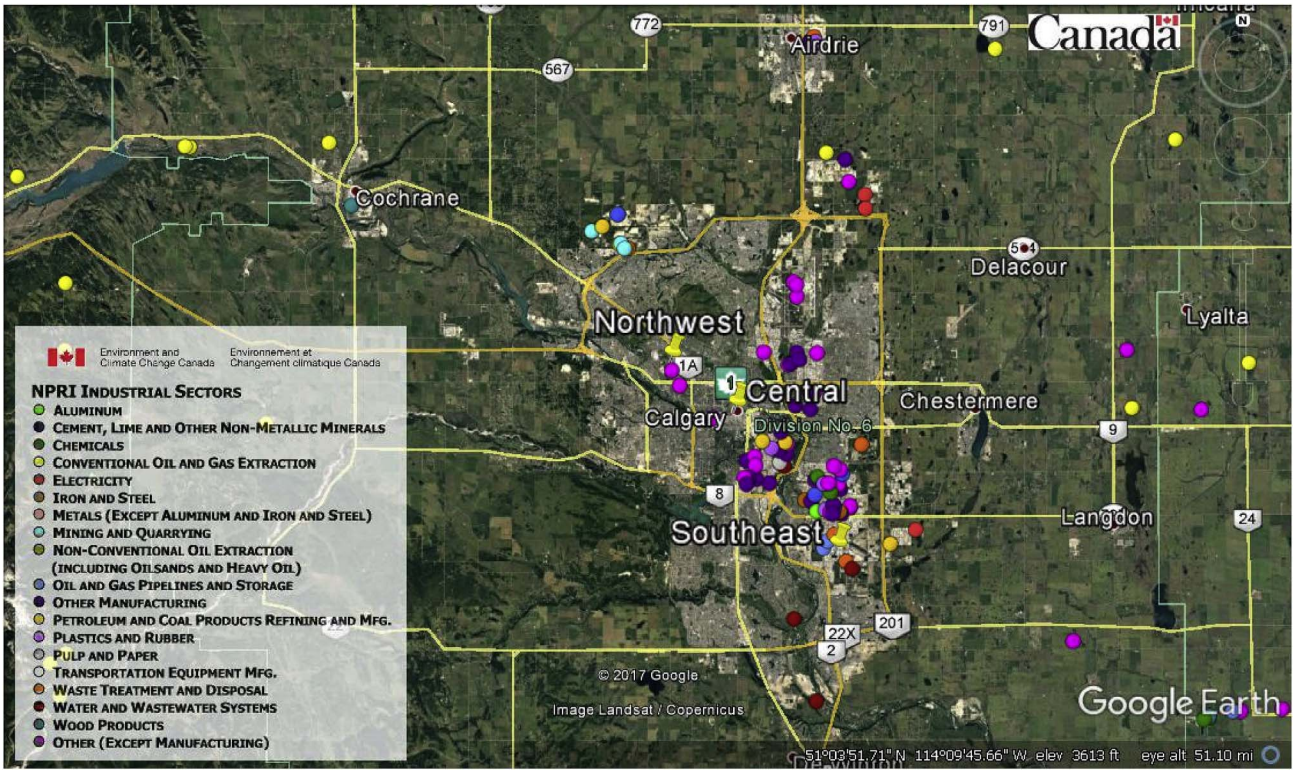

Fig. 1. Locations of three air monitoring stations (yellow stick pins) in Calgary and industries in and surrounding the monitoring stations that report to NPRI during 2015 using Google Earth (Image © 2017 DigitalGlobe © 2017 Google).

(51.0309° N, 114.0089° W, elevation 1036 m) on April 2015 (Fig. S1d). The downtown core of the city lies in the Bow River Valley and surrounding residential areas are elevated to 30–60 m above the floor of the valley. Measurement instruments used for real-time air quality monitoring at Calgary stations are shown in Table S4. Since 1998 hourly concentrations of $PM_{2.5}$ had been measured using Tapered Element Oscillating Microbalances (TEOM @40 °C). From April 2009 the United States Environment Protection Agency's Federal Equivalency Method (FEM) i.e., TEOM coupled with Filter Dynamics Measurement System (TEOM-FDMS) was adopted by Environment Canada to capture semi-volatile organic compounds in $PM_{2.5}$. However, due to challenges and difficulties in maintaining reliable operation of the TEOM-FDMS instrument, the Synchronized Hybrid Ambient Real-time Particulate Monitor (SHARP 5030), a FEM instrument (USEPA, 2015), was adopted at Calgary stations from July 2015. Because of an economic downturn that started in 2014, the study period was selected from January 2014 to December 2016. Seasonal wind roses at the Calgary northwest station were generated for the selected study period (Fig. S3). Prevailing wind directions during all seasons were from the northwest blowing from 55% to 72% of the time, with minor contribution from the south-southeast representing approximately 20% of the time. Meteorological parameters (wind speed and direction) are not monitored at the Calgary central station.

2.2. Data analysis and receptor modeling

Hourly concentration data for criteria air pollutants i.e., nitric oxide (NO), NO_2, NO_X, SO_2, O_3, carbon monoxide (CO) and total hydrocarbon (THC), and meteorological parameters (only available at Calgary northwest) were accessed via the Alberta Environment and Parks air data warehouse (AEP, 2017). Available time integrated 24 h concentrations data for VOCs measured at a frequency of once every 6 days at Calgary central were also accessed for the study period via ECCC (2017a). Both Calgary central and Calgary northwest stations are part of the ECCC's NAPS air monitoring system. Details of sampling, analysis methods and QA/QC guidelines for hourly air pollutants and 24 h VOCs measurements are described elsewhere (Environment Canada, 2004; Wang et al., 2005; Galarneau et al., 2016). Monthly average concentrations of criteria air pollutants, diurnal hourly profiles, and exceedances of CAAQS were investigated to characterize air quality in Calgary. A local source identification tool – conditional bivariate probability functions (CBPF) developed by Uria-telaetxe and Carslaw (2014) – was used for major air pollutants in order to provide information on the nature of local emission sources and potentially identify contributions from different source types through their wind speed dependence for these pollutants. The CBPF includes conditional probability function (CPF) with wind speed as a third variable and it allocates the observed pollutant concentration to a cell defined by ranges of wind direction and wind speed and can be defined as:

$$CBPF_{\Delta\theta,\Delta u} = m_{\Delta\theta,\Delta u|C\geq x}/n_{\Delta\theta,\Delta u}$$

where $m_{\Delta\theta,\Delta u}$ is the number of samples in the wind sector $\Delta\theta$ with wind speed interval Δu having concentration C greater than a threshold value x, and $n_{\Delta\theta,\Delta u}$ is the total number of samples in that wind direction-speed interval. The threshold criterion was set at the highest 25% of the concentrations to define the directionality of local sources.

The United States Environmental Protection Agency's (USEPA's) multivariate receptor model positive matrix factorization (EPA PMF5.0) (USEPA, 2014) was used to determine possible emission sources of measured $PM_{2.5}$ concentrations in the City of Calgary using 24 h concentrations of gaseous pollutants. The model description and data treatment procedures are described in detail in the SI. Data available for seven pollutants (NO, NO_2, NO_X, O_3, CO, THC, CH_4) from January 2014 to December 2016, including 1096 daily (24 h) concentrations, was used for PMF analysis. In this study, several approaches – e.g., plausibility and interpretability of factors, multiple linear regression (MLR), and PMF error estimates –were examined to obtain the optimum number of factor solutions. MLR provides an additional feature for assessing the appropriateness of the chosen number of factors in a PMF

analysis. To quantify the relative contribution of each factor to measured $PM_{2.5}$ concentrations, MLR analysis was performed to regress the measured $PM_{2.5}$ against PMF-derived factor contributions (G matrix).

Different approaches were applied to verify source assignments from the PMF analysis. This involved calculating Spearman's rank correlation coefficients between PMF-derived factor contributions and VOC data in order to investigate their relationship with identified sources and to assist in interpretation of sources. To identify potential local emission sources at the Calgary northwest station, CBPF plots were generated using PMF-derived factor contributions along with wind direction and wind speed. This was not done at the Calgary central station because of the absence of meteorological parameters. However, backward trajectory analysis was conducted at the Calgary central station using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Rolph, 2003) in order to identify the influence of potential long-range sources. Backward trajectory analysis results were plotted using the concentration-weighted trajectory (CWT) method (Seibert et al., 1994; Hsu et al., 2003). Further details on trajectory analysis are provided in the SI.

3. Results and discussion

3.1. Levels of criteria air pollutants

Table 1 shows descriptive statistics of hourly concentrations of criteria air pollutants at Calgary central and Calgary northwest stations over the study period 2014–2016 (monthly profiles of hourly concentrations are presented in Fig. S4). The overall means and median concentrations of $PM_{2.5}$ were similar at both Calgary central (arithmetic mean: 7.7 μg/m^3, median: 6.0 μg/m^3) and Calgary northwest (arithmetic mean: 7.5 μg/m^3, median: 6.0 μg/m^3). Elevated hourly 98th percentile concentrations of $PM_{2.5}$ were observed during summer months particularly in July and August ranging from 30 μg/m^3 to 69 μg/m^3. Several high-$PM_{2.5}$ event days occurred in summer when maximum 24 h concentrations exceeded the CWS of 30 μg/m^3 (e.g., July 17, 2014: 44 μg/m^3; August 25, 2015: 139 μg/m^3), suggesting an influence of wildfire smoke from Alberta and nearby provinces such as British Columbia, and Saskatchewan as well as from the western United States (Fig. S5). Some elevated 98th percentile concentrations were also recorded during winter months e.g., January (30 μg/m^3) and February (31μg/m^3) particularly at Calgary central, when some daily 24 h concentrations (range 25–37 μg/m^3) exceeded the 2015 CAAQS for $PM_{2.5}$ (28 μg/m^3).

Concentrations of NO, NO_2 and NO_X were comparatively higher at Calgary central than Calgary northwest. A clear seasonal trend was observed for NO_2 at both stations, where higher 98th percentile hourly concentrations were observed during winter months from November to March e.g., at Calgary central (45–57 ppb) compared to summer months (23–28 ppb) (Fig. S4). This seasonal effect is likely due to a combination of the role of reactive nitrogen gases (e.g., NO, NO_2) in photochemical O_3 production and subsequent oxidation reactions during summer, which removes these gases from the atmosphere. In addition, temperature prolongs the residence time of these reactive gases during colder months compared to summer. For O_3, 98th percentile hourly concentrations were highest between the months of March through July, with median peaks recorded in April/May (36 ppb) (Fig. S4). The observed spring high pattern is consistent with what others have explained as O_3 inputs originating from the tropospheric reservoir and brought down to the surface from atmospheric boundary layer mixing (Singh et al., 1978; Taylor and Hanson, 1992; Lovett, 1994; Aneja et al., 2000; Steinbacher et al. 2004). An apparent winter-high pattern was observed for CO and THC at Calgary central, where 98th percentile hourly concentrations during November to February ranged from 0.8 ppm to 1.2 ppm for CO and from 2.2 ppm to 2.9 ppm for THC (see Fig. S4).

Diurnal profiles of mean hourly concentrations can aid in understanding temporal behavior of air pollutants and these profiles can be influenced by meteorological dispersion conditions e.g., height of mixing layer, intensity of boundary layer mixing as well as diurnal behavior of emission sources. The average hourly diurnal concentrations profiles of major criteria air pollutants during winter and summer were evaluated at Calgary central and northwest stations for 2014–2016 (Fig. S6). Fig. 2 shows CBPF plots of criteria air pollutants at the Calgary northwest station. These plots represent wind direction locations and wind speed dependence and can be used to tentatively identify the probable influence of emission sources for these air pollutants that are located within 1 h travel time (approximately 15–30 km away depending on wind speed) from the monitoring station.

During winter, NO_X concentration maxima were observed between 8:00 to 10:00 (local time) and 19:00 to 22:00 at both Calgary central and northwest (Fig. S6). Higher NO_X levels in morning hours and increasing levels in evening hours are typical patterns due to daily rush-hour traffic intensity. Calgary central and the northwest station can be influenced by traffic. During summer, NO_X concentration maxima occurred between 7:00 to 9:00 and concentration minima occurred in late afternoon between 18:00 to 19:00. At Calgary northwest, the winter and summer CBPF plots of NO_2 (Fig. 2) indicate near-field sources under low wind speeds suggesting an influence of traffic-related emissions from the adjacent Crowchild Trail NW as well as from residential

Table 1
Descriptive statistics of hourly concentrations of criteria air pollutants at Calgary for the selected study period 2014–2016.

	Calgary central						Calgary northwest					
	Arithmetic mean	Geometric mean	Median	25th percentile	75th percentile	Range (Min–Max)	Arithmetic mean	Geometric mean	Median	25th percentile	75th percentile	Range (Min–Max)
$PM_{2.5}$ (μg/m^3)	7.7	5.6	6.0	3.0	10.0	1.0–185	7.5	5.3	6.0	3.0	9.0	1.0–189
NO (ppb)	11.1	2.7	2.6	0.7	9.7	0.1–331	4.3	1.4	1.2	0.5	3.4	0.1–169
NO_2 (ppb)	17.0	12.6	13.9	7.3	23.9	0.8–74	10.7	7.4	7.7	3.9	14.4	0.1–61
NO_X (ppb)	27.3	16.6	16.5	8.1	32.6	0.2–404	14.7	9.4	9.2	4.8	17.7	0.1–219
O_3 (ppb)	21.2	14.7	21.0	10.0	31.0	1.0–84	26.0	20.8	26.0	15.0	36.0	1.0–90
CO (ppm)	0.21	0.18	0.20	0.1	0.2	0.1–2.4	0.21	0.19	0.20	0.2	0.2	0.1–1.6
THC (ppm)	2.0	2.0	1.9	1.9	2.1	1.6–4.8	1.93	1.92	1.90	1.9	2.0	1.3–3.5
CH_4 (ppm)	2.0	2.0	1.9	1.9	2.0	1.6–4.5	1.92	1.92	1.90	1.9	2.0	1.3–3.2

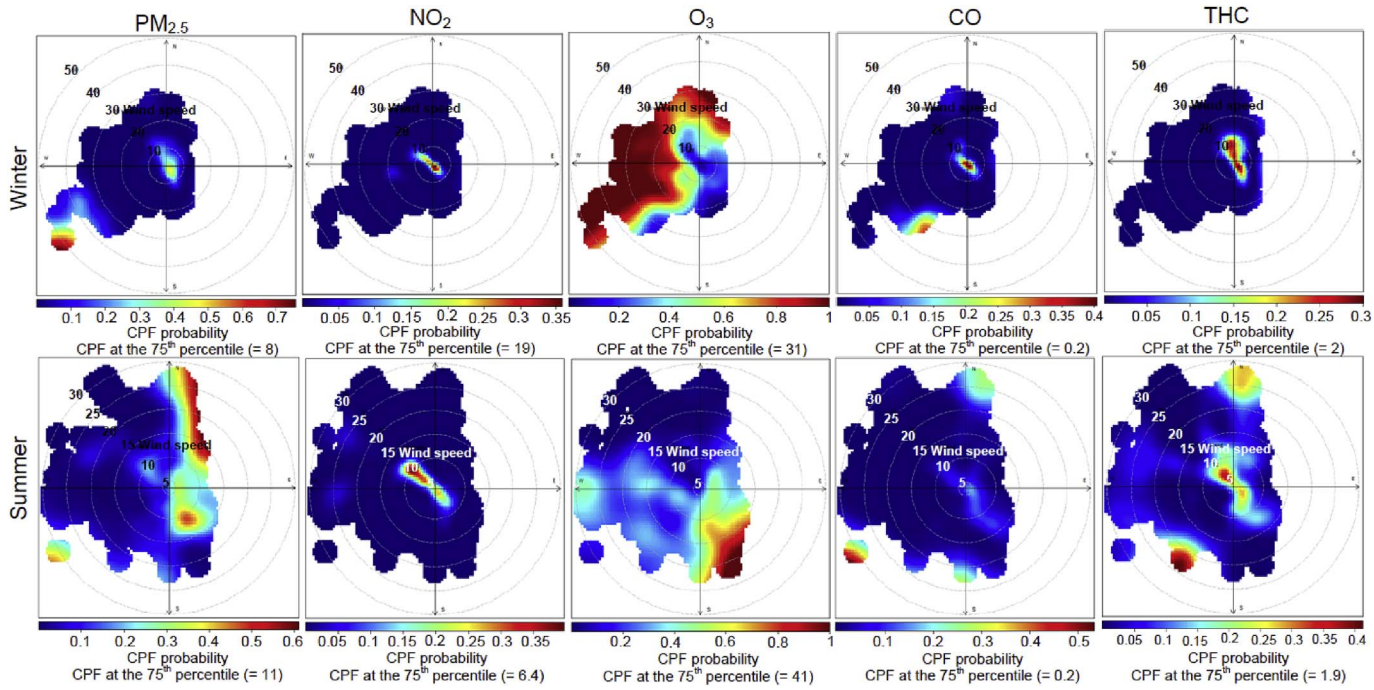

Fig. 2. CBPF plots of major criteria air pollutants at Calgary northwest for the study period 2014–2016.

combustion emissions during winter. Peak concentrations of O_3 were observed during mid-afternoon due to the expected role of daily atmospheric boundary layer mixing and minima occurred during morning hours. The presence of ground level O_3 is strongly influenced by the daily development and dissipation of turbulent mixing within the atmospheric boundary layer (Singh et al., 1978; Zhang and Rao, 1999; Aneja et al., 2000; Steinbacher et al., 2004). Mixing refers to the exchange of ozone-enriched air from higher elevation with lower-ozone air from lower elevations during daily (diurnal) vertical convective mixing within the boundary layer. Daily ozone maxima coincide with when the vertical convective mixing effect is strongest. Vertical convective mixing is at its maximum during mid-afternoon and dissipates during early to late evening. When depth of the boundary layer increases during mid-morning hours, ozone suspended aloft is mixed downward to the earth's surface and surface concentrations increase. Once the atmospheric boundary layer mixing ceases starting in early to late evening, and during the night hours, ozone surface concentrations decrease due to scavenging by chemical species such as NO. The CBPF plot for O_3 (Fig. 2) indicated dominant north, northwest and southwest directions during winter and southeast directions during summer months.

The diurnal hourly average pattern for $PM_{2.5}$ (Fig. S6) exhibited maxima between 7:00 to 9:00 during summer and between 10:00 to 12:00 (at Calgary central) and 21:00 to 23:00 (both stations) during winter, while minima occurred during afternoon hours (15:00 to 17:00). The winter CBPF plot for $PM_{2.5}$ (Fig. 2) suggests traffic (adjacent Crowchild Trail NW) as a moderate source. While the summer CBPF plot for $PM_{2.5}$ pointed north, northeast, and southeast as probable major source directions. Like NO_X and $PM_{2.5}$, the diurnal concentration patterns for CO (Fig. S6) showed maxima between 9:00 to 11:00 and 20:00 to 24:00 during winter and between 7:00 to 9:00 during summer and are likely associated with combustion sources including traffic and commercial and residential heating. The CBPF plot for CO (Fig. 2) suggested local (traffic from Crowchild Trail NW) and southwest as dominant source directions during winter and north as moderate influence during summer. For THC no clear diurnal pattern was observed (Fig. S6). The winter CBPF plot for THC (Fig. 2) suggested traffic (adjacent Crowchild Trail NW), while the summer CBPF plot suggested traffic and a dominant influence from the southwest and a moderate one from the north.

To evaluate the levels of particulate air pollution in Calgary, temporal profiles of 24 h $PM_{2.5}$ concentrations were examined using

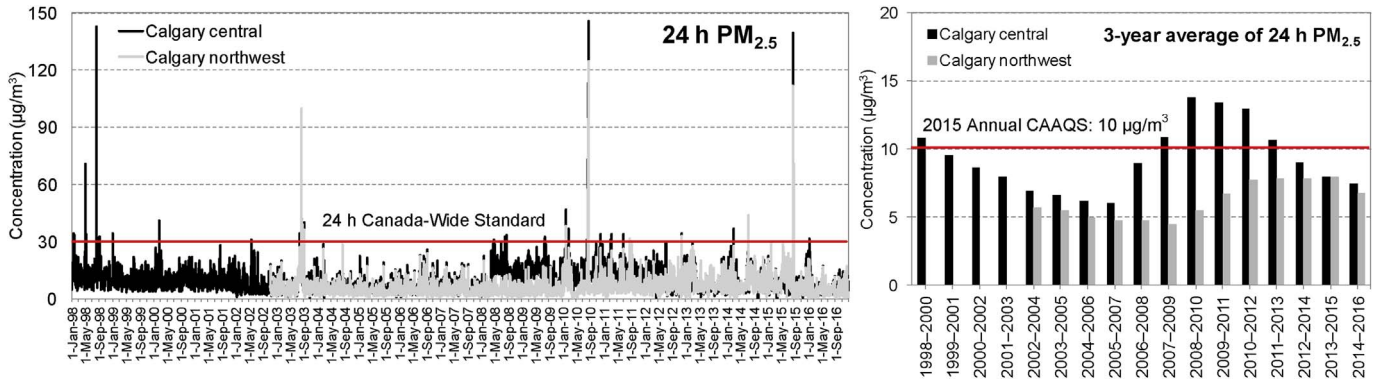

Fig. 3. Time series of 24 h $PM_{2.5}$ (left) and the 3-year average of the annual average $PM_{2.5}$ concentrations (right) at Calgary central station (3 downtown locations over the period) and Calgary northwest station for 2014–2016.

available data at Calgary central station, which included 3 downtown locations over the period 1998 to 2016, and at Calgary northwest station for the period 2003 to 2016 (Fig. 3). Most 24 h exceedances above the CWS of 30 μg/m^3 were recorded during summer months, suggesting that forest fire events play a major role in the exceedances of $PM_{2.5}$ standard. During summer months several forest fire episodes occurred in Alberta and nearby provinces and territories such as British Columbia (August 19, 2010; July 17, 2014), Saskatchewan (June 30, 2015) and from the western United States (August 25, 2015) (CWFIS, 2015). Numerous 24 h exceedances were also observed during winter months and are likely associated with emission sources such as traffic, commercial/residential heating coupled with winter time stable weather conditions. Fig. 3 also shows the 3-year average of the annual average of the daily 24 h $PM_{2.5}$ concentrations. At the downtown locations (Calgary central), levels exceeded the 2015 annual CAAQS of 10 μg/m^3 during 2000 and from 2009 to 2013, while no exceedances occurred for 2004–2016 at Calgary northwest. The observed exceedances at the downtown Calgary locations over 2009–2013 may, in part, be associated with changes in the monitoring methods (i.e., TEOM-FDMS and Beta Attenuation Monitor-BAM 1020). Levels went down at Calgary central from 2014 to 2016 (Fig. 3), which may, in part, be due to relocation of the central monitoring location from Calgary central 2 to Calgary-Inglewood (4.8 km away) and changes in the monitoring method over this time (i.e., replacement of the TEOM-FDMS/BAM 1020 with a SHARP 5030).

3.2. $PM_{2.5}$ source identification in Calgary

The PMF model was applied in this study to gaseous pollutants data and a 5-factor solution was optimized based on MLR analysis and PMF error estimates. $PM_{2.5}$ mass was then apportioned using MLR to the 5 PMF-resolved sources at Calgary central and northwest stations for 2014–2016. Model input data statistics, performance criteria and error estimates are shown in the SI (Tables S5–S7). The source profile from the base run and time series plot of daily contributions of sources are shown in Fig. 4. The average contribution (in percent) of PMF-derived sources to $PM_{2.5}$ for the study period is shown in Fig. 5 and seasonal average contributions are presented in Table S8. Winter and summertime CBPF plots of the five identified sources are shown in Fig. 6 (CBPF plots for the whole study period are shown in SI Fig. S7). The spatial distribution of winter and summertime CWT values for the contribution of two long-range sources e.g., secondary aerosol and combustion is depicted in Fig. 7. Potential long-range source regions of the combustion source during specific wildfire smoke intrusion event days in Calgary during the study period 2014–2016 are shown in Fig. S8 based on 72-h back trajectories. CWT plots for an O_3-rich source are shown in Fig. S9. Table S9 shows the MLR model summary and coefficients of sources and predictor variables. The regression relationship between measured $PM_{2.5}$ and five identified source factors showed a moderate association with a MLR-derived adjusted R^2 of 0.40 and 0.48 at Calgary central and Calgary northwest, respectively, suggesting that the observed $PM_{2.5}$ concentrations were moderately represented with the resolved five sources.

3.2.1. Factor 1 - secondary aerosol

Factor 1 was interpreted as secondary aerosol and it was the most dominant source at both Calgary central and Calgary northwest contributing 54% and 42% of the $PM_{2.5}$ mass concentration on average, respectively (Fig. 5). It was characterized by a notable abundance of THC and CH_4 (57% of pollutants sum) explaining 44% and 45% of the variation, respectively. Some small mass fractions of CO (18%) and O_3 (15%) were also present in this factor. THC is a broad family group, dominated by large background CH_4 concentrations along with aromatic hydrocarbons (i.e., containing one or more benzene rings) and aliphatic (non-aromatic) hydrocarbons. THC can be emitted from a variety of sources including vegetation, traffic, petroleum and chemical industries, biomass and natural gas combustion and other fugitive sources.

In Calgary background $PM_{2.5}$ concentrations may arise from local sources and background sources that bring $PM_{2.5}$ into the area via regional transport from natural (e.g., biogenic) and anthropogenic origins (e.g., upstream oil and gas development surrounding Calgary and throughout Alberta). Secondary inorganic (e.g., sulfate, nitrate) and organic aerosol (SOA) may also contribute to $PM_{2.5}$ in Calgary. In Alberta, background regional sulfate is found in relative abundance due to oil and gas production (e.g., natural and sour gas extraction, flaring and processing), other industrial emissions like coal- and gas-fired industrial boilers for power generation and other non-specific industrial

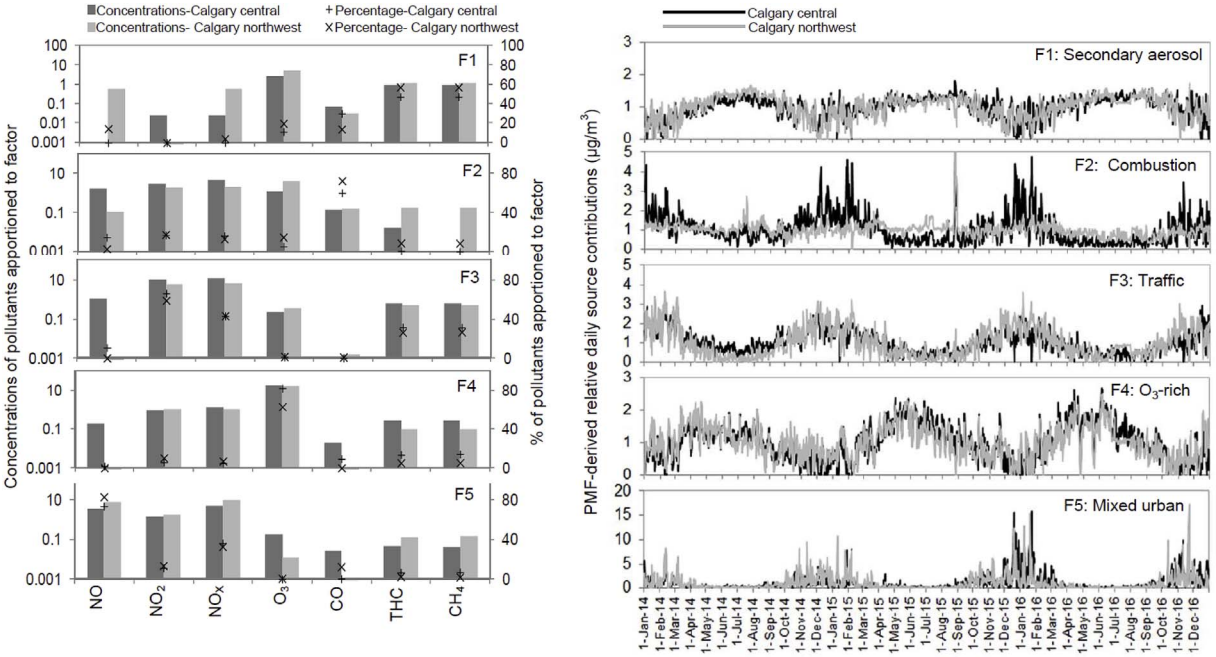

Fig. 4. Source profile and daily contributions of five identified sources at Calgary central and northwest stations for 2014–2016.

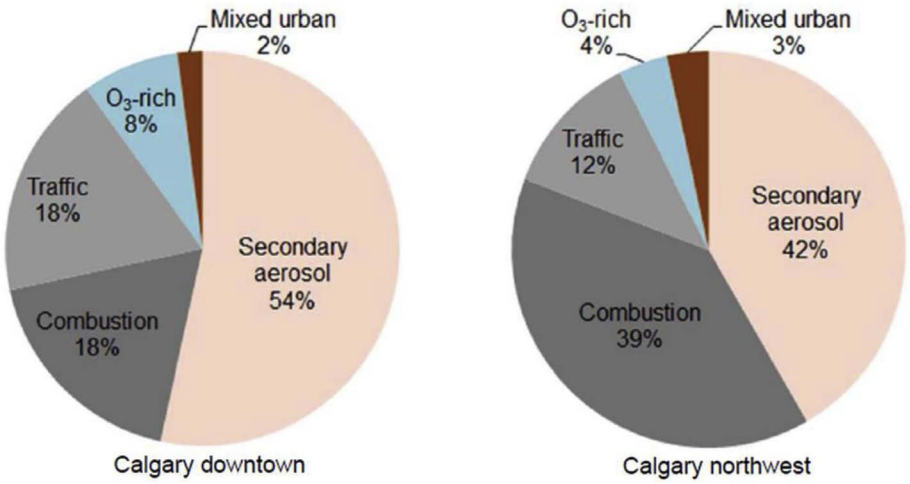

Fig. 5. Average contribution (in percent) of PMF-derived sources to $PM_{2.5}$ for the study period 2014–2016.

sources (Schulz and Kindzierski, 2001). In an earlier study during 1985–1995 (Cheng et al., 1998), the sulfate content in ambient $PM_{2.5}$ mass was found at ~11% in Calgary and Edmonton, consistent with that observed in rural-remote and rural-influenced sites (11%–40%) in Alberta (Cheng et al., 2000).

In general, secondary aerosol formation is enhanced during summer month photochemical activity (Turpin and Huntzicker, 1995) and can be linked to various local and long-range transport of anthropogenic and natural emissions such as traffic exhaust, fossil fuel combustion, biomass burning and biogenic emissions (Hoyle et al., 2011; Skyllakou et al., 2014). The time series plot of this factor showed a clear seasonality (Fig. 4) with higher contributions in spring and summer months compared to fall and winter (Table S8). This factor was positively correlated with known tracers of biogenic emissions i.e., isoprene and α-pinene, suggesting a notable contribution of biogenic activities (Table S10). The annual contribution of this factor showed significant variation (Kruskal-Wallis test, $p < 0.0001$) over the study period 2014–2016 at both monitoring stations at Calgary (Tables S11, S12).

Both winter and summer CBPF plots of this factor (Fig. 6) revealed northwest and southwest (only wintertime) as dominant source directions under high wind speeds and moderate influence from the north and southeast under low wind speeds. In terms of possible sources, several local industries are located within 10 km northwest and also the Inter pipeline extraction plant and Wildcat Hills gas plant in Cochrane (located 29 km and 38 km, respectively from the northwest monitoring station) may be partially associated with this factor. The time-series plot (Fig. 4) also indicated some peaks during summer and winter months. In terms of other sources, summertime wildfire events (e.g., August 25, 2015) and wintertime traffic and biomass burning activities within and surrounding Calgary may also be partially associated with this factor. From backward trajectory analysis (Fig. 7), the winter CWT plot indicated higher potential long-range contributions to the immediate north and south of Calgary, and from west and southwest Alberta, British Columbia and Western United States (long-range), suggesting that residential wood-burning and open wood burning (e.g., agricultural slash burning) may have been occurring during winter. The

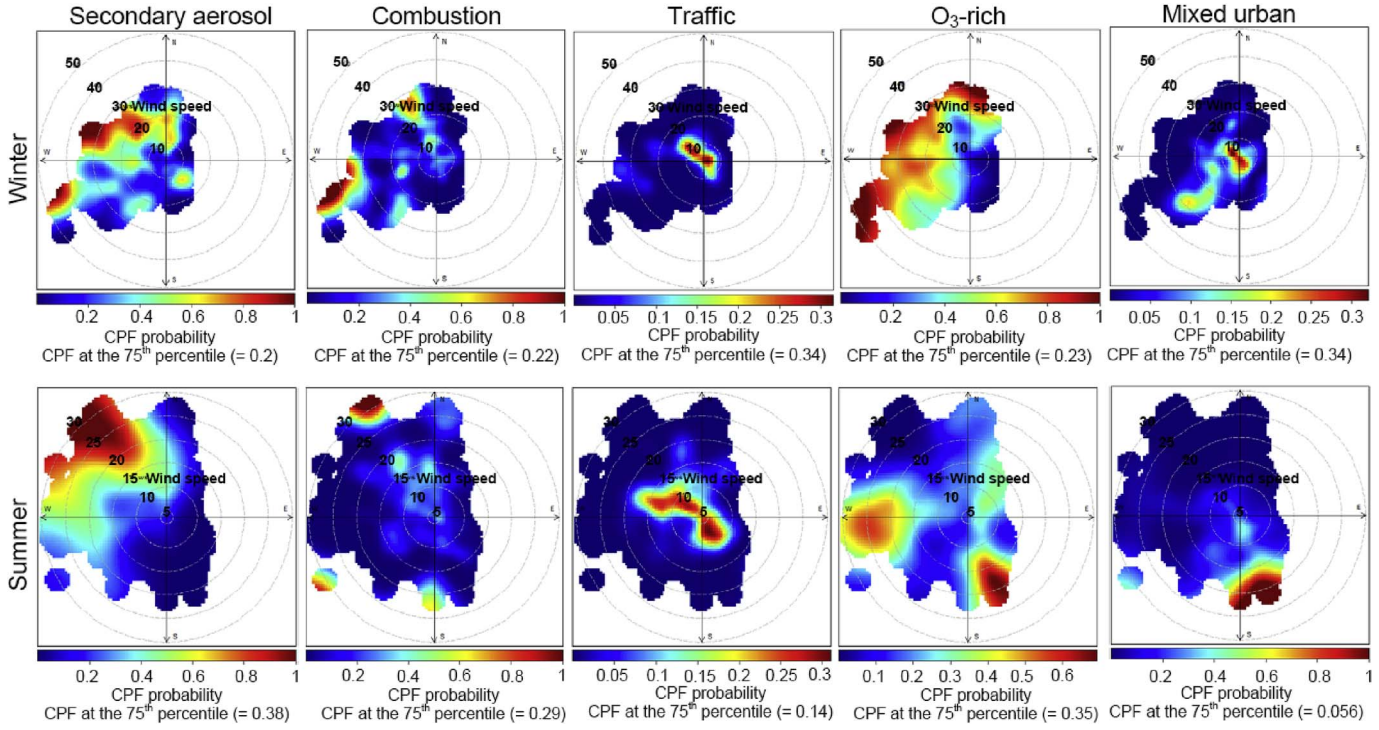

Fig. 6. Winter and summertime CBPF plots of PMF-derived sources at Calgary northwest for 2014–2016.

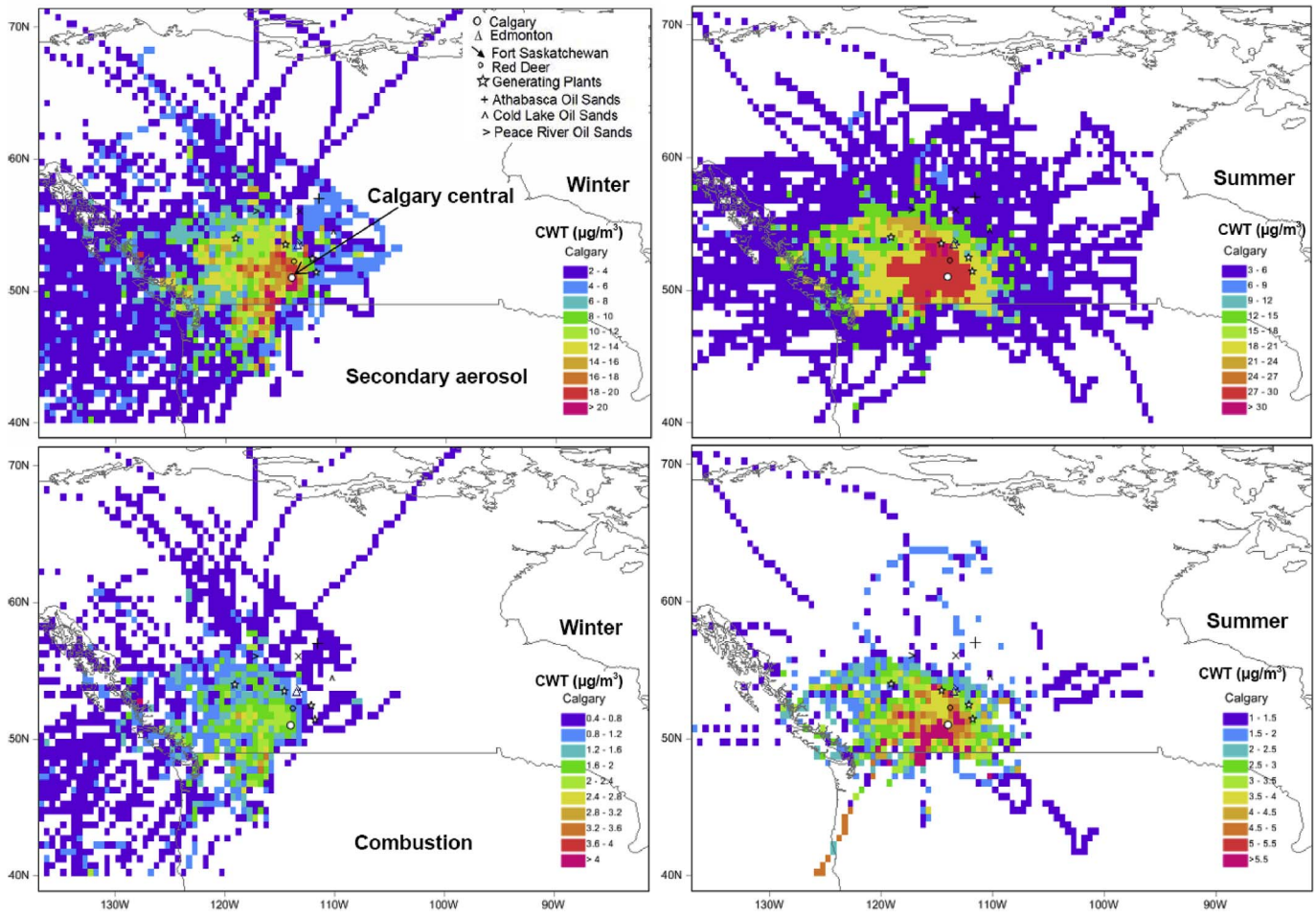

Fig. 7. Winter and summer CWT values of secondary aerosol and combustion sources at Calgary central for 2014–2016.

summer CWT plot indicated potential source regions surrounding Calgary, central and southern Alberta, British Columbia and United States (long range). Therefore, factor 1 was best represented as secondary aerosol source. It is acknowledged that due to lack of secondary tracer compounds data (e.g., sulfate, nitrate and organic aerosols), clear identification of secondary inorganic and organic aerosols were not possible in this study. However, the observed contribution of secondary aerosol source in Calgary was comparable to secondary inorganic (32%) and organic aerosol (27%) sources identified in a recent source apportionment study using $PM_{2.5}$ chemical species in an urban area of Edmonton, Alberta (located 300 km from Calgary) for the period 2009–2015 (Bari and Kindzierski, 2016b).

3.2.2. Factor 2 - combustion

Factor 2 was characterized by high levels of CO (up to 73% of pollutants sum) explaining 54% of the variation along with some contributions of NO_2, NO_X, THC, CH_4 (representing 13%–23% of the explained variation). CO is released to the atmosphere primarily from incomplete combustion of carbon-containing fuels such as natural gas, gasoline, oil and wood. The time-series plot of daily source contributions of this factor (Fig. 4) showed numerous peak concentrations (e.g., July 17, 2014; June 30, 2015; August 25, 2015) during summer months, suggesting that this factor is associated with wildfire smoke emissions that occurred in Alberta and other western provinces such as British Columbia, Saskatchewan and in Western United States during the period 2014–2016. Several peaks were also observed during winter months indicating a possible influence of emissions from commercial and residential heating. As reported previously, according to CRAZ 2008 emission inventories more than 10,000 tonnes/year of CO along with VOCs (> 1500 tonnes/year), NO_X (> 5000 tonnes/year) and $PM_{2.5}$ (~2000 tonnes/year) are released in the air of CRAZ region and the City of Calgary from commercial and residential heating sector (Table S2). Combustion of natural gas for commercial/residential heating and wood stove/fireplace use among residences within the city and surrounding rural Calgary, respectively during winter appears to be common. Contributions of this factor showed significant correlations with typical tracers for biomass combustion (Liu et al., 2008) e.g., ethylene, ethane, acetylene, propylene, 1,3-butadiene, benzene (Spearman's rank coefficient, $r = 0.74–0.89$, $p < 0.01$) and tracers for natural gas combustion e.g., ethane, propane, butane, isobutane ($r = 0.71–0.81$, $p < 0.01$) (Table S10). Significant year to year variation (Kruskal-Wallis test, $p < 0.0001$) was observed in contributions of this factor at both Calgary stations (Tables S11, S12).

The winter CBPF plot of this factor (Fig. 6) indicated northwest and southwest as probable dominant directions for local sources. The winter CWT plot (Fig. 7) indicated potential moderate source regions immediately west and northwest of Calgary, central and southern Alberta, United States suggesting the possible influence of residential wood-burning and open wood burning (e.g., agricultural slash burning) that may have been occurring during winter (Bari and Kindzierski, 2016b). The summer CWT plot indicated an influence of wildfire smoke coming from British Columbia, northern and southern Alberta, Saskatchewan and the United States (Figs. 7 and S8). Therefore, factor 2 was assigned as combustion. As expected the contribution of this factor to $PM_{2.5}$ was higher at the residential monitoring station Calgary northwest (39%) compared to Calgary central (18%). Combustion, particularly biomass burning, was also identified as an important contributor to $PM_{2.5}$ mass in other urban areas in Alberta – e.g., Edmonton (13%), Red Deer (11%)

and Fort McMurray (8%) (Bari and Kindzierski, 2016a,b; 2017a,c).

3.2.3. Factor 3 - traffic

Factor 3 was represented by high concentrations of traffic-related gaseous pollutants i.e., NO_2, and NO_X explaining up to 57% and 44% of the variation, respectively. Some contributions of NO, THC and CH_4 (8%–30% of their total mass) were also observed. This factor was well correlated with vehicle exhaust-related VOCs e.g., benzene, ethylene, acetylene, propylene, 1,3-butadiene, 1-butene ($r = 0.59–0.78$, $p < 0.01$), moderately correlated with vehicle evaporative-related VOCs e.g., *n*-butane, isobutene, *n*-pentane, isopentane, ($r = 0.45–0.66$, $p < 0.01$) and positively correlated with liquid/unburned gasoline and diesel fuel-related species e.g., C5 and C6 alkanes (i.e., pentanes and hexanes) and other vehicle-related species e.g., ethylbenzene, isomers of xylenes, ethyltoluenes, trimethylbenzenes (Harley et al., 1992; Schauer et al., 1999; Watson et al., 2001) (Table S10). This factor showed a clear seasonality with high contributions in fall and winter compared to other seasons (Table S8). The annual contributions of this factor showed significant variation in Calgary central ($p < 0.0001$), while no variation was observed at Calgary northwest ($p = 0.332$). At Calgary northwest, both winter and summertime CBPF plots of this factor (Fig. 6) revealed southeast, northwest and west (only in summer) as dominant source directions, suggesting traffic influences. Therefore, factor 3 was interpreted as a traffic source and the contribution of this factor to measured $PM_{2.5}$ mass was higher at Calgary central (18.4%) compared to Calgary northwest (11.9%) (Fig. 5).

3.2.4. Factor 4 - O_3-rich

Factor 4 was represented by a high abundance of O_3 explaining up to 78% of the variation along with some associations with THC and CH_4 (each representing 23% of the explained variation). This factor was interpreted as a general O_3-rich source and it contributed 7.6% and 4.1% to the $PM_{2.5}$ mass concentration on average at Calgary central and Calgary northwest, respectively (Fig. 5). This factor showed negative correlation with most VOC species except chloromethane, where a significant correlation was observed ($r = 0.47$, $p < 0.01$). Chloromethane is an ozone-depleting VOC, released into the atmosphere predominantly from natural sources, while it can also be emitted from some anthropogenic sources such as fossil fuel combustion, waste incineration and industrial processes (McCulloch et al., 1999). The time series plot of this factor indicated a notable seasonality particularly at Calgary central (Fig. 4) with higher contributions in spring and summer months compared to fall and winter. No significant variation in annual contributions was observed at both Calgary stations (Tables S11, S12).

The winter CBPF plot (Fig. 6) at Calgary northwest indicated probable dominant local source directions from the northwest and southwest under high wind speed (> 20 km/h), while the summer CBPF plot revealed strong influences from the west and southeast directions. As reported previously, several potential local industries located within 10 to 40 km northwest and 10 to 30 west and east/southeast (Table S3) may partially contribute to this factor. The winter and summertime CWT plots (Fig. S9) indicated potential long-range contributions from the north/northwest and southern Alberta, British Columbia and the United States. Industry emissions (upstream oil and gas in Alberta and mining, pulp mill and wood processing in British Columbia) are possible long-range sources for this factor. Using real-time monitoring data, similar to the method used here, an 'O_3-rich' factor was also identified in other studies conducted in Rochester, New York (Kasumba et al., 2009), Prague, Czech Republic (Thimmaiah et al., 2009), Kuala Lumpur, Malaysia (Khan et al., 2015) and in Red Deer, Alberta (Bari and Kindzierski, 2017a).

3.2.5. Factor 5 - mixed urban

Factor 5 was small and characterized by high levels of NO, and NO_X explaining 78% and 38% of the variation, respectively and also showed some association with NO_2 and CO (14% and 16% of the variation, respectively). At Calgary central daily contributions of this factor showed significant associations with aromatics, alkanes, alkenes, and tetrachloroethylene (a tracer for dry cleaning, Wallace, 1989) (Table S10). A clear winter-high trend was observed (Fig. 4). At Calgary northwest, the winter and summer CBPF plots (Fig. 6) showed northwest and southeast (during winter) and southeast (during summer) as probable dominant directions suggesting both traffic and local industry as possible sources. Therefore, factor 5 was assigned as a mixed urban source and it only contributed 2.0% and 3.4% to measured $PM_{2.5}$ mass concentration on average at Calgary central and Calgary northwest, respectively.

4. Conclusion

An exploratory study of air quality and sources of $PM_{2.5}$ concentrations was undertaken in the City of Calgary, Alberta for the period 2014 to 2016. Summer $PM_{2.5}$ concentrations were relatively higher compared to other seasons. The 3-year averages of the annual average 24 h $PM_{2.5}$ concentrations (8.1–8.9 μg/m^3) were below the 2015 annual CAAQS value of 10 μg/m^3. Observed summertime 24 h exceedances of the 2015 CAAQS at Calgary stations were primarily due to the influence of wildfire smoke intrusion events, while elevated winter levels and 24 h exceedances were likely due to a combination of traffic, industry, commercial/residential heating emissions coupled with wintertime stable weather conditions. PMF analysis identified secondary aerosol as a dominant contributor to $PM_{2.5}$ mass at Calgary central (54%) and Calgary northwest (42%) followed by combustion (18%, 39%), traffic (18%, 12%), an O_3-rich source (8%, 4%), and a minor mixed urban source (2%, 3%), respectively.

A number of limitations exist in this study. Due to the lack of chemical speciation data in $PM_{2.5}$, we could not identify and separate the contribution of several potentially important sources that influence urban particulate matter in Alberta – such as secondary inorganic (sulfate, nitrate) and organic aerosol, biogenic emissions, agriculture, construction, road dust and specific industry-related emission sources (e.g., cement industry, manufacturing/metallurgy), as well as potential inputs from different long-range industrial emission sources throughout Alberta, e.g. upstream oil and gas activities. We acknowledge that uncertainty exists in source apportionment results obtained using gaseous pollutants data. $PM_{2.5}$ chemical speciation monitoring would be needed to better understand the contribution of these sources to $PM_{2.5}$ in a large urban area like Calgary. Notwithstanding these limitations, our findings offer preliminary information about different emission sources that influence $PM_{2.5}$ levels in the City of Calgary; and this information can support policy makers in developing appropriate air quality management initiatives for particulate matter pollution if needed.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.apr.2017.11.014.

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