



Concentrations, sources and human health risk of inhalation exposure to air toxics in Edmonton, Canada



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HIGHLIGHTS

- Health risk and sources of ambient air toxics assessed in Edmonton for 2009–2013.
- Carcinogenic risks of air toxics were below acceptable and/or tolerable risks.
- Traffic and background/secondary organic aerosol contributed >60% to total HAPs.
- Findings are useful for developing risk management and controlling air toxics.

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ABSTRACT

With concern about levels of air pollutants in recent years in the Capital Region of Alberta, an investigation of ambient concentrations, sources and potential human health risk of hazardous air pollutants (HAPs) or air toxics was undertaken in the City of Edmonton over a 5-year period (2009–2013). Mean concentrations of individual HAPs in ambient air including volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs) and trace metals ranged from 0.04 to 1.73 $\mu\text{g}/\text{m}^3$, 0.01–0.54 ng/m^3 , and 0.05–3.58 ng/m^3 , respectively. Concentrations of benzene, naphthalene, benzo(a)pyrene (BaP), arsenic, manganese and nickel were far below respective annual Alberta Ambient Air Quality Objectives. Carcinogenic and non-carcinogenic risk of air toxics were also compared with risk levels recommended by regulatory agencies. Positive matrix factorization identified six air toxics sources with traffic as the dominant contributor to total HAPs (4.33 $\mu\text{g}/\text{m}^3$, 42%), followed by background/secondary organic aerosol (SOA) (1.92 $\mu\text{g}/\text{m}^3$, 25%), fossil fuel combustion (0.92 $\mu\text{g}/\text{m}^3$, 11%). On high particulate air pollution event days, local traffic was identified as the major contributor to total HAPs compared to background/SOA and fossil fuel combustion. Carcinogenic risk values of traffic, background/SOA and metals industry emissions were above the USEPA acceptable level (1×10^{-6}), but below a tolerable risk (1×10^{-4}) and Alberta benchmark (1×10^{-5}). These findings offer useful preliminary information about current ambient air toxics levels, dominant sources and their potential risk to public health; and this information can support policy makers in the development of appropriate control strategies if required.

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1. Introduction

Exposure to hazardous air pollutants (HAPs), commonly known as air toxics is of concern due to potential carcinogenic and other adverse health effects including respiratory, neurological, reproductive and developmental effects (Delfino, 2002; Windham et al., 2006; USEPA, 2012). Under the United States 1990 Clean Air Act Amendments, 187 air toxics have been identified that warrant

specific attention and long-term monitoring, and 30 HAPs have been defined as urban air toxics which pose the greatest potential threat to public health in urban areas (USEPA, 2016a,b). In Canada several of these 187 HAPs are treated as ‘toxic substances’ by Health Canada under Canadian Environmental Protection Act (CEPA) 1999.

Characterization of ambient air toxics is beneficial for understanding sources and human health risks of HAPs based on inhalation exposure. HAPs may consist of a variety of organic and inorganic species including volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs) and metals. They can be emitted from a wide range of natural (e.g., forest fires) and

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anthropogenic sources including traffic, fossil fuel industries, residential and commercial heating, evaporation processes, paints and solvent industries (Wu et al., 2007; Logue et al., 2009). Strum and Scheffe (2015) recently analyzed ambient air observations of 27 HAPs across the United States in order to characterize national concentrations and risks of urban air toxics. In Canada Galarneau et al. (2016) investigated ambient concentrations of 63 air toxics under the National Air Pollution Surveillance (NAPS) program. However, to the best of our knowledge limited source and risk apportionment studies (e.g., Logue et al., 2009; Wu et al., 2007; Kavouras et al., 2015) have been carried out worldwide so far to investigate sources of measured ambient HAPs out of the lists of 187 USEPA air toxics and their associated risk to public health in urban areas.

Edmonton (area 684 km², population of 877,926 in 2014) is the second largest urban area and the centre of the Capital Region of Alberta and is located on the North Saskatchewan River, about 300 km north of Calgary and about 400 km east of the foothills of the Canadian Rockies. Edmonton has a relatively humid continental climate with wide fluctuations in seasonal temperatures. In general, cold winter and mild summer seasons last from November 15 to March 15 and May 15 to September 15, respectively. Average prevailing wind directions are west-northwesterly and early morning surface inversions are common throughout the whole year in Alberta with deeper and stronger inversions observed in November through January (Myrick et al., 1994; AESRD, 2014). The city is surrounded by numerous fossil fuel industries (8 km east: Strathcona petroleum refineries; ~30 km northeast: Alberta's industrial heartland in Fort Saskatchewan-several chemical and petrochemical industries, manufacturing industries e.g., steel mill; ~70 km west: coal-fired generating plants) and agricultural farming lands (Myrick et al., 1994; Schulz and Kindziarski, 2001; AESRD, 2014) (Supplemental Information-SI, Fig. S1). National Pollutant Release Inventory (NPRI) (Environment Canada, 2016a) annual releases of VOCs, PAHs, and some trace metals (e.g., As, Ni) to the air from major industrial facilities/operations in and surrounding Edmonton and in Alberta (Fig. S2, Tables S1, S2) for 2009–2013 are shown in the SI. In previous studies traffic, industrial activities and biomass burning were found as dominant emission sources of ambient and outdoor VOCs (McCarthy et al., 2013; Bari et al., 2015a) and trace metals in submicron particles (PM₁) (Bari et al., 2015b) in Edmonton and the surrounding region.

A recent media release stated that Alberta was on track to have worst air quality in Canada (National Post, 2015a). In addition, a Canadian physicians group claimed that Edmonton experienced higher levels of harmful air pollutants compared to Toronto and blamed coal-fired power generating plants for Edmonton's poor air quality (National Post, 2015b). This raises questions about current levels of air pollutants of public health concerns in Edmonton and the surrounding region. A study was therefore carried out to characterize selected air toxics of public health concerns for the most recent years in Edmonton. The objectives of this study were to better understand ambient levels of air toxics, determine their potential local and long-range sources in Edmonton and evaluate their risk to public health.

2. Methodology

2.1. Ambient sampling and chemical analysis

The study was undertaken using available HAPs data collected from air monitoring stations in Edmonton. Under Environment Canada's NAPS program, Alberta Environment and Parks (AEP) routinely monitors criteria air pollutants including HAPs in Edmonton air monitoring stations (Fig. S1). The study was carried

out using publicly available HAPs data for a 5-year period (2009–2013) accessed via Environment Canada (2016b). Ambient 24 h data for 33 hazardous volatile organic compounds (VOCs) were taken from Edmonton central station. VOC samples were collected at a frequency of once every 6 days using 6-L evacuated Summa canisters (Scientific Instrumental Specialists, Inc.) and analyzed by gas chromatograph-mass spectrometer (GC-MS) at Environment Canada's lab in Ottawa. Details of VOCs sampling and analysis methods are described elsewhere (Wang et al., 2005; Galarneau et al., 2016). Data for 9 PAHs including 7 carcinogenic ones were taken from Edmonton McIntyre station (collected 1-in-6 day frequency) for the study period except in 2012 (collected 1-in-6 day frequency at Edmonton east station). Data for 9 hazardous trace metals in PM_{2.5} were obtained from Edmonton McIntyre station. Ambient 24 h integrated PM_{2.5} samples were collected gravimetrically using Partisol 2300 sequential speciation sampler (Thermo Fisher Scientific, USA) on a 1-in-3 day and trace metals were analyzed by inductively-coupled plasma mass spectrometry (ICP-MS). Further details of the analytical methods and quality assurance/quality control (QA/QC) procedures are described elsewhere (Celo et al., 2010; Dabek-Zlotorzynska et al., 2011; McCarthy et al., 2013). Winter and summer wind roses at McIntyre station were generated for the 5-year study period (2009–2013) (Fig. S3) based on historical hourly meteorological observations via the AEP airdata warehouse (AEP, 2016a). Twenty-four hour concentration data for secondary pollutants (SO₄²⁻, NO₃⁻, NH₄⁺), organic carbon (OC), elemental carbon (EC) and other organic (e.g., levoglucosan, arabitol) and elemental (e.g., Ba, Cu, V, Zn) markers were also accessed for the study period for Edmonton McIntyre station via Environment Canada (2016b).

2.2. Source apportionment method

To identify and apportion possible emission sources of ambient HAPs in Edmonton, the United States Environmental Protection Agency (USEPA)'s multivariate receptor model positive matrix factorization (EPA PMF5.0) (USEPA, 2014a) was used. Details of the model description, data treatment procedure, uncertainty or error estimation methods are presented in the SI. The input file included a 5-year dataset (2009–2013) consisting of 205 daily (24 h) samples (excluding one sample due to outlier), where all measured HAPs data (VOCs, PAHs, trace metals) were available on selected days. Out of 51 measured HAPs, 37 species including 19 VOCs, 9 PAHs and 9 trace metals were selected for PMF analysis based on their higher frequency of detection (specifically, those HAPs with at least 50% of the data above the method detection limit (MDL)) and using selected HAPs as source-specific markers (refer to Table S3). Multiple linear regression (MLR) analysis was conducted to regress total measured HAPs against PMF-derived factor contributions in order to determine the relative contribution of each of the identified sources to total measured HAPs. Different statistical approaches were applied to verify the PMF-resolved source assignments. Data for criteria air pollutants and meteorological parameters were used to investigate their relationship with identified sources by calculating Spearman's rank correlation coefficient for each source. To better identify and characterize potential local emission sources, a conditional bivariate probability function (CBPF) (Uria-telegaetxe and Carslaw, 2014) was calculated using PMF-derived factor contributions by coupling conditional probability function (CPF) with bivariate polar plots in order to show hidden contributions from different source types through their wind speed dependence. The impact of potential long-range sources was also investigated using the concentration-weighted trajectory (CWT)

model for backward trajectory analysis (Seibert et al., 1994; Hsu et al., 2003). Further details on trajectory analysis are provided in the SI.

2.3. Human health risk assessment

To evaluate the potential public health risk posed by inhalation exposure to air toxics, a screening human health risk assessment was performed. In this study, all available HAPs including VOCs, PAHs and trace metals with known cancer and non-cancer toxicity values were considered. For assessing risks associated with long-term (chronic) and short-term (acute) inhalation exposures, dose-response criteria of the USEPA's Office of Air Quality Planning and Standards (OAQPS) were used to screen ambient concentrations of HAPs in Edmonton based on the 5-year dataset. For chronic exposure, carcinogenic and non-carcinogenic risks were evaluated using EPA inhalation unit risk (IUR , $(\mu\text{g}/\text{m}^3)^{-1}$) and reference concentrations ($RfCs$, mg/m^3) associated with the concentration for each HAPs species (USEPA, 2014b, 2015). For acute exposure, available United States Agency for Toxic Substances and Disease Registry (ATSDR) minimal risk levels (MRLs) and California EPA reference exposure levels (RELs) were used.

Health risk from inhalation exposure to PMF-derived HAPs sources was estimated by summing carcinogenic and non-carcinogenic risks of all available risk-posing species in a particular source. We applied a 'point estimate approach' that has been used in several studies (Wu et al., 2009; Li et al., 2015; Khan et al., 2016; Bari and Kindzierski, 2016). In this study, exposure concentration (EC) was calculated using the following equation according to USEPA's Superfund program (USEPA, 1991, 2009):

$$EC_{ij} = C_{ij} \times \frac{ET \times EF \times ED}{AT}$$

where EC_{ij} and C_{ij} are the exposure concentration and concentration of the j th species from the i th source ($\mu\text{g}/\text{m}^3$), respectively, ET : air exposure time (24 h/day), EF : exposure frequency (350 days/year, assuming 15 vacation days per year), ED : exposure duration (24 years for adults), AT : average time (for carcinogens, $70 \text{ yrs} \times 365 \text{ days/yr} \times 24 \text{ h/day}$; for non-carcinogens, AT : $ED \times 365 \text{ days} \times 24 \text{ hrs/day}$). The cancer and non-cancer risk from exposure to the i th source was estimated as the sum of cancer and non-cancer risks of all available n risk-posing HAPs species in PMF-derived profiles.

$$\text{Carcinogenic risk}_i = \sum_{j=1}^n EC_{ij} \times IUR_j$$

$$\text{Non-carcinogenic risk}_i = \sum_{j=1}^n \frac{EC_{ij}}{RfC_j}$$

where, IUR_j and RfC_j are the inhalation unit risk and the reference concentration for the j th species, respectively. The excess cancer risk range recommended by the USEPA (2009) for public health protection is a one in a million (1×10^{-6}) acceptable risk level to one in a thousand (1×10^{-4}) tolerable risk level. Provincial regulatory agencies across Canada have varying guidance of acceptable cancer risk, with Alberta, British Columbia, and the Atlantic provinces accepting an incremental lifetime cancer risk of 1 in 100,000 (1×10^{-5}) (Health Canada, 2004; Kindzierski et al., 2011). A non-cancer risk is represented by hazard index (HI). USEPA (2009) states that if $HI < 1$, there is no appreciable risk of adverse health effects, while $HI > 1$ indicates a chance of non-cancer effects occurring.

3. Results and discussion

3.1. Ambient levels of HAPs and health risk posed by inhalation exposure

Table 1 shows ambient concentrations of detected HAPs in Edmonton for 2009–2013 and comparison with the ATSDR risk levels and USEPA reference concentrations and risk levels (detailed summary statistics of detected HAPs and data quality of all measured HAPs are shown in Table S3). Out of 51 measured HAPs, descriptive statistics is presented for 38 species based on a higher data completeness of at least 30% of samples above the detection limit. Average and geometric mean concentrations of major VOCs e.g., individual BTEX (benzene, toluene, ethylbenzene and xylenes) ranged from 0.28 to $1.73 \mu\text{g}/\text{m}^3$ with maximum of up to $16.21 \mu\text{g}/\text{m}^3$. Benzene concentrations were far below the annual Alberta Ambient Air Quality Objective (AAAQO) (AEP, 2016b) of $3 \mu\text{g}/\text{m}^3$. Concentrations of *n*-Hexane (median $0.56 \mu\text{g}/\text{m}^3$, range 0.06– $4.15 \mu\text{g}/\text{m}^3$) were about 2- to 3-fold lower at Edmonton downtown compared to Edmonton east petrochemical industrial site (data not shown), consistent with previous studies (Cheng et al., 1997; Bari et al., 2015a). For long-lived halogenated VOCs, elevated levels were observed for chloromethane, dichloromethane (DCM), 1,4-Dichlorobenzene (*p*-DCB), tetrachloroethene (PERC) and carbontetrachloride (CCl_4). Naphthalene showed low levels (average $0.11 \mu\text{g}/\text{m}^3$) well below the annual AAAQO of $3 \mu\text{g}/\text{m}^3$. The most carcinogenic PAHs, i.e., benzo(*a*)pyrene (BaP) (average $0.06 \text{ ng}/\text{m}^3$) was also found below the annual AAAQO of $0.3 \text{ ng}/\text{m}^3$. Retene, a typical marker for biomass combustion (Ramdahl, 1983) showed elevated levels (median $0.29 \mu\text{g}/\text{m}^3$, range 0.03– $8.35 \text{ ng}/\text{m}^3$), possibly suggesting an influence of biomass smoke emissions, for example from wintertime heating (wood stoves, fireplaces) and summertime forest fires. Concentrations of potential toxic trace metals e.g., arsenic (As) (geomean $0.16 \text{ ng}/\text{m}^3$), nickel (Ni) (geomean $0.35 \text{ ng}/\text{m}^3$) were also far below their annual AAAQO of $10 \text{ ng}/\text{m}^3$ and $50 \text{ ng}/\text{m}^3$, respectively (AEP, 2016b).

For health risk screening, arithmetic and geometric mean concentrations of selected HAPs were compared to non-cancer (RfC) and cancer (IUR) risk levels for potential chronic end-points, and maximum concentrations were compared to acute MRL and REL values. Concentrations were also compared with cancer benchmark targets used for Alberta (1 in 100,000). Geometric and arithmetic means of all HAPs were well below all USEPA screening risk criteria and cancer benchmark for Alberta except for some PAHs e.g., BaP, BbF and DahA, where mean concentrations were just above or meeting the benchmark targets. The carcinogenic and non-carcinogenic risks of HAPs species via inhalation exposure are shown in Fig. S4. Carcinogenic risks of most HAPs exposure were less than the acceptable level of 1×10^{-6} , except for benzene, 1,3-butadiene, carbon tetrachloride and naphthalene, where median and mean concentrations were just above or meeting the acceptable level but below the tolerable risk (1×10^{-4}). The 95th percentile carcinogenic risks of some toxic trace metals e.g., As, Cr and Co were just above and/or meeting the acceptable level, while integrated risk from all detected HAPs were below a tolerable risk of 1×10^{-4} . In Alberta, these types of potential lifetime exposures are deemed acceptable if it increases a person's chance of developing cancer by 1 in 100,000 or less (Health Canada, 2004; Kindzierski et al., 2011). This level appears quite reasonable when compared to the average background risk of all cancers (estimated at 33,890 in 100,000 over a lifetime) that Albertans carry (Kindzierski et al., 2011). For non-carcinogenic risk, geometric and arithmetic means and 95th percentile values of HQ of all HAPs and HI were well below the safe level of concern ($= 1$).

Table 1Edmonton HAPs of concern for 2009–2013 compared to ATSDR risk levels and USEPA reference concentrations and risk levels ($\mu\text{g}/\text{m}^3$).

HAPs	HAP No.	IARC WOE	Edmonton, Alberta				Chronic						Acute	
			Ambient concentrations				Non-cancer		Cancer				MRL	REL
			Units	Average	Gmean	Max	RfC $\mu\text{g}/\text{m}^3$	Source	EPA WOE	IUR 1 in 10^{-6} 1/	Source	Benchmark* 1 in 10^{-5} ($\mu\text{g}/\text{m}^3$)		
Benzene	15	1	$\mu\text{g}/\text{m}^3$	0.72	0.63	3.31	30	IRIS	CH	7.8E-06	IRIS	1.28	29	1300
Toluene	152	3	$\mu\text{g}/\text{m}^3$	1.73	1.43	11.96	5000	IRIS	InI	—	—	—	3800	37000
Ethyl benzene	77	2B	$\mu\text{g}/\text{m}^3$	0.35	0.28	3.14	1000	IRIS	D	2.5E-06	CAL	4.0	22000	—
Xylenes (mixed)	169	3	$\mu\text{g}/\text{m}^3$	1.64	1.31	16.21	100	IRIS	InI	—	—	—	8700	22000
n-Hexane	95	—	$\mu\text{g}/\text{m}^3$	0.74	0.58	4.15	700	IRIS	InI	—	—	—	—	—
1,3-Butadiene	23	1	$\mu\text{g}/\text{m}^3$	0.09	0.07	0.43	2	IRIS	CH	0.00003	IRIS	0.33	220	—
Styrene	146	2B	$\mu\text{g}/\text{m}^3$	0.07	0.05	0.57	1000	IRIS	—	—	—	—	21000	21000
Chloroform	39	2B	$\mu\text{g}/\text{m}^3$	0.09	0.09	0.25	98	ATSDR	LH	—	—	—	490	150
Chloromethane	106	3	$\mu\text{g}/\text{m}^3$	1.18	1.17	1.92	90	IRIS	InI	—	—	—	1000	—
Bromomethane	105	—	$\mu\text{g}/\text{m}^3$	0.07	0.06	0.20	5	IRIS	D	—	—	—	190	3900
Dichloromethane (DCM)	116	2B	$\mu\text{g}/\text{m}^3$	0.42	0.37	3.68	600	IRIS	LH	1E-08	IRIS	1000	2100	14000
1,4-Dichlorobenzene (p-DCB)	53	2B	$\mu\text{g}/\text{m}^3$	0.41	0.15	33.07	800	IRIS	C	1.1E-05	CAL	0.91	12000	—
1,2 Dichloroethane (DCA)	81	2B	$\mu\text{g}/\text{m}^3$	0.07	0.06	0.28	2400	ATSDR	B2	2.6E-05	IRIS	0.38	—	—
1,1,1-Trichloroethane (TCA)	107	—	$\mu\text{g}/\text{m}^3$	0.05	0.04	0.22	5000	IRIS	InI	—	—	—	11000	68000
Trichloroethylene (TCE)	159	2A	$\mu\text{g}/\text{m}^3$	0.07	0.052	0.56	2	IRIS	CH	4.1E-06	IRIS	2.44	11000	—
Tetrachloroethene (PERC)	150	2A	$\mu\text{g}/\text{m}^3$	0.26	0.19	1.55	40	IRIS	LH	2.6E-07	IRIS	38.46	1400	20000
Carbon tetrachloride (CCl_4)	29	2B	$\mu\text{g}/\text{m}^3$	0.54	0.53	0.72	100	IRIS	LH	6E-06	IRIS	1.67	—	1900
2,2,4-Trimethylpentane (TMP)	164	—	$\mu\text{g}/\text{m}^3$	0.55	0.45	3.49	—	—	—	—	—	—	—	—
Naphthalene	119	2B	$\mu\text{g}/\text{m}^3$	0.11	0.09	0.58	3	IRIS	C	3.4E-05	CAL	0.29	—	—
Benzo(a)pyrene (BaP)	187	1	ng/m^3	0.06	0.03	1.54	—	—	B2	0.0011	CAL	0.01	—	—
Benzo(a)anthracene (BaA)	187	2B	ng/m^3	0.08	0.04	1.54	—	—	B2	0.00011	CAL	0.09	—	—
Chrysene (Chr)	187	2B	ng/m^3	0.13	0.08	1.61	—	—	B2	1.1E-05	CAL	0.91	—	—
Benzo(b)fluoranthene (BbF)	187	2B	ng/m^3	0.15	0.09	2.33	—	—	B2	0.00011	CAL	0.09	—	—
Benzo(k)fluoranthene (BkF)	187	2B	ng/m^3	0.05	0.029	0.84	—	—	B2	0.00011	CAL	0.09	—	—
Dibenz(a,h)anthracene (DahA)	187	2A	ng/m^3	0.01	0.011	0.19	—	—	B2	0.0012	CAL	0.01	—	—
Indeno(1,2,3-cd)pyrene (IP)	187	2B	ng/m^3	0.08	0.05	1.51	—	—	B2	0.00011	CAL	0.09	—	—
Retene	187	—	ng/m^3	0.54	0.32	8.35	—	—	—	—	—	—	—	—
Benzo(g,h,i)perylene (BgHiP)	187	—	ng/m^3	0.13	0.08	1.89	—	—	—	—	—	—	—	—
Arsenic (As)	174	1	ng/m^3	0.28	0.16	12.21	0.015	CAL	A	0.0043	—	2.3E-03	—	—
Cadmium (Cd)	176	1	ng/m^3	0.07	0.05	0.82	0.01	ATSDR	B1	0.0018	—	5.6E-03	—	—
Cobalt (Co)	178	2B	ng/m^3	0.16	0.05	4.80	0.1	ATSDR	—	0.009	—	1.1E-03	—	—
Chromium (Cr)	177	1	ng/m^3	0.62	0.44	2.93	0.1	IRIS	CH	0.012	—	8.3E-04	—	—
Manganese (Mn)	183	—	ng/m^3	3.58	2.50	12.48	0.3	ATSDR	D	—	—	—	—	—
Nickel (Ni)	186	1	ng/m^3	0.57	0.35	4.11	0.09	ATSDR	A	0.00024	—	4.2E-02	—	—
Lead (Pb)	182	2B	ng/m^3	0.58	0.38	6.76	0.15	OAQPS	B2	—	—	—	—	—
Antimony (Sb)	173	—	ng/m^3	0.21	0.17	1.25	—	—	—	—	—	—	—	—
Selenium (Se)	189	3	ng/m^3	0.09	0.07	0.46	20	CAL	D	—	—	—	—	—

IRIS = Integrated Risk Information System; ATSDR = US Agency for Toxic Substances and Disease Registry; CA L = California EPA; OAQPS = USEPA's Office of Air Quality Planning and Standards; IARC WOE = weight-of-evidence for carcinogenicity in humans (1 - carcinogenic; 2A - probably carcinogenic; 2B - possibly carcinogenic; 3 - not classifiable). EPA WOE (2005 and 1986 Guidelines) = weight of evidence for carcinogenicity: CH - carcinogenic to humans, A - human carcinogen; B1 - probable carcinogen, limited human evidence; B2 - probable carcinogen, sufficient evidence in animals; C - possible human carcinogen; D - not classifiable, InI - inadequate information to assess carcinogenic potential; MRL = ATSDR minimum risk levels for no adverse effects for 1–14-day exposures. REL = California EPA reference exposure level for no adverse effects. Most, but not all, RELs are for 1-h exposures; —, not available, *Cancer benchmark used in Alberta, British Columbia, and the Atlantic provinces except in Ontario and Quebec (Health Canada, 2004).

3.2. HAPs source characterization

In this study, the plausibility and interpretability of solutions with five to ten factors were checked and a 6-factor PMF solution was optimized to best represent HAPs sources in Edmonton for 2009–2013. The six identified sources included traffic, background/secondary organic aerosol (SOA), fossil fuel combustion, biomass burning, metals industry and mixed urban. Model input data statistics and regression diagnostics are shown in the SI (Tables S4, S5).

A summary of PMF analysis and diagnostics of error estimations for 5 to 7 factors are shown in Table S6. Increasing or decreasing the number of factors showed splitting or combined sources, respectively and did not appear to be physically meaningful. For example, a 7-factor solution yielded an additional As-rich factor and an 8-factor solution yielded two additional As-rich and p-DCB-rich sources. For a 5-factor solution, biomass burning and mixed urban factors were combined to a single factor. Bootstrapping (BS) analysis showed that results were generally stable in the 6-factor

solution compared to 5- and 7-factor solutions, where all 6 factors mapped to a base factor in 100% of runs except for the biomass burning factor (mapped 88% of runs) (Table S6). In bootstrapping with displacement (BS-DISP) error analysis, 91% and 87% of cases were accepted and there were zero and one swaps in best fit for 5- and 6-factor solutions, respectively indicating stable solutions. While for a 7-factor solution only 52% cases were accepted and some swaps were present, indicating some uncertainty with the solution. Therefore, in this study a 6-factor solution was chosen based on the BS and BS-DISP results and clear interpretability of the factors.

Fig. 1 shows source profiles i.e., chemical composition of emission sources for each of the six identified source factors from the base and the bootstrap runs (average contributions from the base run in the parentheses). A time series plot of daily contributions of sources is shown in Fig. S5. Variability in the concentration of sources from the bootstrap run is shown in Fig. S6 and correlation analysis is reported in Table 2. CBPF plots of the six identified sources in Edmonton are shown in Fig. 2 for visualization of potential contribution sources within 1-h travel time. The spatial distribution of winter CWT values in Edmonton is presented in the SI (Figs. S7–S9) for visualization of potential contribution of long-range sources. Table S7 shows the MLR model summary and coefficients of sources and predictor variables. The regression relationship between modeled and measured concentrations of total HAPs showed a good fit (adjusted $R^2 = 0.88$) with modeled resolved sources explaining 88% ($p < 0.0001$) of the variance in measured HAPs in Edmonton (Fig. S10). This suggests that the observed total HAPs concentrations were represented quite well with the resolved 6 factors. From using PMF-MLR approach, the sum of the contributions of the 6 identified sources accounted for 86% of the measured HAPs on average, while remaining 14% of the measured HAPs was unexplained (Fig. 1, Table S7).

3.2.1. Traffic

Factor 1 was identified as the largest source at Edmonton accounting for 42% ($4.33 \mu\text{g}/\text{m}^3$) of the measured HAPs on average. It was characterized by high concentrations of vehicle exhaust related tracer species e.g., benzene, toluene, 1,3-butadiene, ethylbenzene, xylene isomers (Watson et al., 2001; Schauer et al., 1999) explaining from 44% to 64% of the variation. High contributions of hexane, styrene, 2,2,4-trimethylpentane and naphthalene (43%–58% of the explained variation) along with some association of trace metals (As, Co, Sb, Mn, Ni) and PAH species (BaA, Chr, BbF, BkF, IP, BghiP) explaining 11–30% and 13–19% of the variation, respectively were also found in this factor. The toluene to benzene (T/B) ratio of 1.5–3.0 has been generally used as an indicator of vehicle emissions (Miller et al., 2010). In this factor the T/B ratio of 3.1 was observed indicating the influence of traffic emissions. As, Co, Sb, Mn, Ni can be also linked to non-exhaust traffic emissions from brake/tire wear, and road and pavement erosion (Sternbeck et al., 2002; Lough et al., 2005; Thorpe and Harrison, 2008). Contributions of this factor showed significant correlations with traffic related components e.g., NO_3^- , OC, EC, Ba, Cu and Zn (Table 2). The time series plot of daily contributions (Fig. S5) showed clear seasonality with about two-fold higher contributions in winter ($5.72 \mu\text{g}/\text{m}^3$) and fall ($5.31 \mu\text{g}/\text{m}^3$) compared to spring ($2.60 \mu\text{g}/\text{m}^3$) and summer ($3.47 \mu\text{g}/\text{m}^3$) months. Several factors such as stable atmospheric conditions (low temperature and wind speed, surface inversions), cold-start emissions and increased car idling can contribute to higher concentrations during winter. The CBPF plot for this factor (Fig. 2) revealed west and southwest as dominant directions under high wind speed and less dominant southerly and easterly directions under low wind speed, suggesting an influence of local roadways. The presence of the local highway Whitemud Drive

(Highway 14 and Highway 2 bypass, Fig. S11) located within less than 5 km from the Edmonton McIntyre station are likely associated with this factor. The results agree well with previous source apportionment studies carried out in Edmonton for $\text{PM}_{2.5}$ and VOCs (Jeong et al., 2011; McCarthy et al., 2013; Bari et al., 2015a; Bari and Kindzierski, 2016). Therefore, factor 1 can be best represented as a traffic source.

3.2.2. Background/secondary organic aerosol (SOA)

Factor 2 was the second largest source in Edmonton, contributing 25% ($1.92 \mu\text{g}/\text{m}^3$) to the total measured HAPs on average. The predominant HAPs species found in this factor were halogenated VOCs e.g., chloroform, chloromethane, DCM, *p*-DCB, DCA, TCA, TCE, PERC, CCl_4 (explaining from 31% to 60% of the variation) and showed notable association with naphthalene, BaP, DahA (38%, 20%, 46% of their total mass, respectively). Some contributions of As (21%), Cr (22%), Sb (22%) and Se (72%) were also found. Most halogenated VOCs (e.g., TCA, TCE, *p*-DCB) are released to the atmosphere from industrial and commercial sources (e.g., wastewater treatment plants, solvents, dry cleaning and/or pesticides) (Harkov et al., 1987; Wallace et al., 1987), though some (e.g., chloromethane) occur naturally (Koppmann, 2007). These halogenated species tend to have longer atmospheric lifetimes and can be considered global background pollutants. This factor showed significant correlation with biogenic tracer species (Tanner and Zielinska, 1994; Chebbi and Carlier, 1996; Burshtein et al., 2011) e.g., arabitol, mannitol, isoprene, α -pinene, oxalate ($r = 0.31$ – 0.96 , $p < 0.01$, Table 2) with higher contributions being observed in spring and summer months (Fig. S5). Therefore, factor 2 was interpreted as secondary organic aerosol (SOA).

SOA can be linked to various local and long-range transport of anthropogenic and natural emissions such as traffic, fossil fuel combustion primarily due to evaporation during fossil fuel extraction, processing and transportation, biomass burning and biogenic emissions (Hoyle et al., 2011; Skyllakou et al., 2014; Liggiio et al., 2016). Some associations with As, Cr, Sb and Se may possibly link to vehicular and industrial emissions leading to the formation of secondary aerosol (Bari and Kindzierski, 2016). The CBPF plot of this factor (Fig. 2) showed the highest concentrations with high wind speeds ($>20 \text{ km/h}$) originated from the east, southeast and southwest directions. The presence of local industries to the east, south-east (Sherwood Park), and southwest (Devon, Nisku, Leduc) located within 30 km from downtown Edmonton are likely associated with this factor. Seasonal CWT plots suggest potential regional sources in northern and southern Alberta, British Columbia, Saskatchewan and the U.S. regions (Fig. S7), similarly reported in other studies (Jeong et al., 2011; Bari and Kindzierski, 2016).

3.2.3. Fossil fuel combustion

Factor 3 was represented by high abundances of Pb and Cd explaining 70% and 42% of the variation, respectively and also showed association with As, Se, and Cr (28%, 18% and 9% of the variation, respectively). Some mass fractions of benzene (23%), hexane (20%) and 1,3-butadiene (19%) were also present in this factor. As, Pb, Cd, Se, Cr are typically emitted from different industries including metallurgy, manufacturing, chemical, and petrochemical industries and coal- and gas-fired industrial boilers, power plants (Pacyna, 1987; Goodarzi et al., 2008). Benzene, hexane, 1,3-butadiene, 1,2-dichloroethane and trichloroethylene can be emitted from oil and gas extraction and processing, and petrochemical industry (Lin et al., 2004; Xie and Berkowitz, 2006; Liu et al., 2008). This factor showed significant correlation with secondary pollutants e.g., SO_4^{2-} , NO_3^- , NH_4^+ ($r = 0.46$ – 0.62 , $p < 0.01$) and with EC, V, Zn, and K ($r = 0.28$ – 0.42 , $p < 0.01$, Table 2). A notable seasonality was observed with higher contributions in

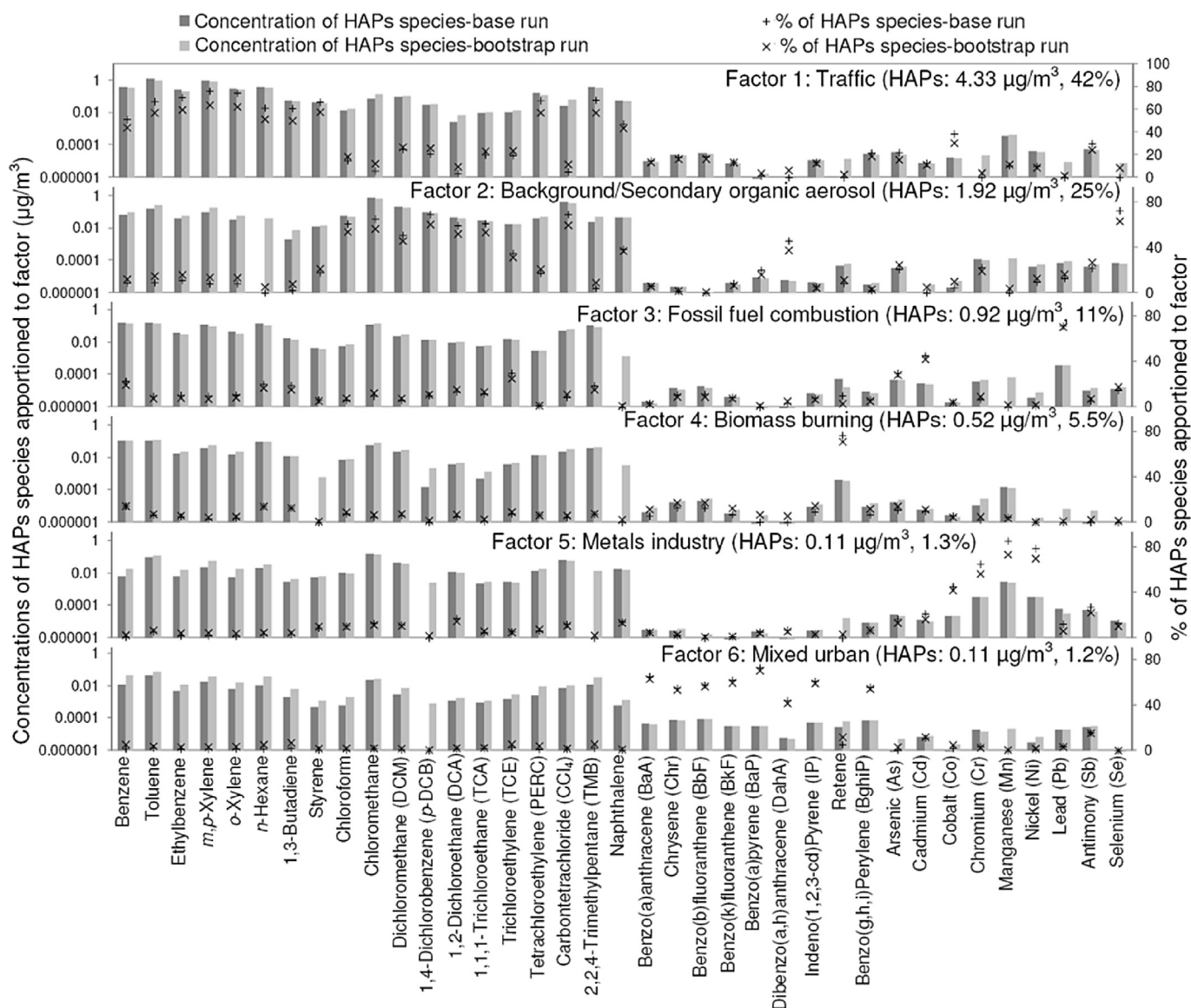


Fig. 1. PMF-derived source profiles (concentration and % of components apporportioned to factor in left and right axis, respectively) and average contributions (in the parentheses) of individual sources to measured total HAPs in Edmonton for 2009–2013.

winter ($1.44 \mu\text{g}/\text{m}^3$) and spring ($1.08 \mu\text{g}/\text{m}^3$) months than in summer ($0.53 \mu\text{g}/\text{m}^3$) and fall ($0.60 \mu\text{g}/\text{m}^3$). For local point source impacts, the CBPF plot of this factor (Fig. 2) indicated the northeast and south as dominant source directions, while lesser influence was found from northwest and west directions. The presence of Strathcona industries (e.g., two major oil refineries Imperial Oil and Suncor Energy 5 km east) and Alberta's industrial heartland in Fort Saskatchewan (within 30 km north and northeast of Edmonton McIntyre station) where a number of petrochemical plants, upgraders, refineries, other industries including agriculture and manufacturing (e.g., steel mill) exist, are likely associated with this factor. Similar local fossil fuel combustion source directions were found in a Bari and Kindzierski (2016) Edmonton $\text{PM}_{2.5}$ source apportionment study for secondary sulfate, secondary nitrate, refinery and metallurgy factors.

From backward trajectory analysis, seasonal CWT plot (Fig. S8) indicated potential source regions to the immediate east/northeast (Strathcona industries area and industrial heartland located in Fort Saskatchewan area), and to the south in Alberta, where upstream oil and gas industries exist. Background regional sulfate found in

abundance throughout Alberta from oil and gas extraction and production/processing activities is a plausible source (Figs. S1, S12). It is noteworthy that coal-fired generating plants located about 70 km west of Edmonton in the Wabamun Lake area showed less contribution to this factor (e.g., CWT values $< 0.6 \mu\text{g}/\text{m}^3$ in summer, Fig. S8), while a generating plant 220 km southeast of Edmonton tended to show greater contribution. Therefore, factor 3 was designated as fossil fuel combustion and the contribution of this factor was 11% ($0.92 \mu\text{g}/\text{m}^3$) of total measured HAPs in Edmonton.

3.2.4. Biomass burning

The most abundant species found in factor 4 was retene explaining 71% of the variation along with some contributions of Chr, BbF, IP, benzene, hexane, 1,3-butadiene, Cd and As (12%–17% of the explained variation). On average this factor contributed 5.5% ($0.52 \mu\text{g}/\text{m}^3$) to the measured HAPs in Edmonton. Retene is a typical candidate tracer for softwood burning smoke (Ramdahl, 1983; Bari et al., 2009). This factor was substantially correlated with known biomass smoke tracers e.g., levoglucosan, mannosan and EC ($r = 0.41$ – 0.56) and also correlated with other biomass-related

Table 2
Correlation (r = Spearman's rank coefficient) of PMF-derived factors with major PM_{2.5} components measured in Edmonton.

	SO ₄ ²⁻	NO ₃ ⁻	NH ₄ ⁺	OC	EC	Levo-glucosan	Mann-osan	Arabitol	Mannitol	Oxalate	Isoprene	α -Pinene	Ba	Cu	Fe	V	Zn	K	Cl ⁻
F1	-0.12	0.19**	0.02	0.27**	0.45**	0.28**	0.32**	0.22**	0.001	0.09	0.45**	0.58**	0.52**	0.34**	0.12	0.03	0.21**	0.11	0.18*
F2	-0.01	-0.48**	-0.15*	0.02	-0.19*	-0.13	-0.11	0.31**	0.96**	0.31**	0.35**	0.32**	0.06	-0.11	0.07	-0.01	-0.31**	-0.31**	-0.28**
F3	0.55**	0.46**	0.62**	0.04	0.30**	0.27**	0.21**	-0.04	-0.18*	0.27*	-0.24**	-0.19*	-0.09	-0.04	0.07	0.28**	0.42**	0.41**	0.19*
F4	-0.09	0.29**	0.04	0.11	0.41**	0.55**	0.56**	0.17*	-0.06	-0.10	0.20*	0.20*	0.25**	0.20**	0.01	-0.05	0.29**	0.35**	0.23**
F5	0.10	0.12	0.06	-0.05	0.37**	0.08	0.14	0.001	-0.07	0.21**	0.16	0.08	0.47**	0.70**	0.70**	0.29**	0.39**	0.32**	0.20*
F6	0.05	0.56**	0.21**	0.13	0.50**	0.47**	0.49**	-0.09	-0.31*	-0.17*	-0.08	-0.02	0.29**	0.27**	-0.002	0.04	0.40**	0.39**	0.39**

F1: Traffic; F2: Background/soa; F3: Fossil fuel combustion; F4: Biomass burning; F5: Metals industry; F6: Mixed urban.

**correlation is significant at $p = 0.01$, *correlation is significant at $p = 0.05$, representative correlations are marked as bold.

species e.g., arabitol, isoprene, α -pinene, K⁺, Zn, Cl⁻, Cu ($r = 0.17$ – 0.35) (Table 2). This factor showed clear seasonality with high concentrations during winter months (Fig. S5), suggesting the influence of wintertime residential heating (e.g., from wood stoves/fireplaces) and agricultural slash burning (Bari and Kindzierski, 2016). The CBPF plot of this factor (Fig. 2) indicated highest concentrations in Edmonton when high winds blew from north and northwest directions. Some high concentrations were also observed in spring and summer months, suggesting the influence of long range transport of wildfires smoke. Winter CWT plot (Fig. S9) suggested potential source regions in central Alberta (e.g., Parkland County immediately west of Edmonton and south towards Red Deer), while summer CWT plot suggested a more distant influence of biomass smoke from northern Alberta, British Columbia, Saskatchewan and the U.S. regions. Therefore, factor 4 was assigned as a biomass burning source.

3.2.5. Metals industry

The predominant HAPs species found in factor 5 were Mn, Ni, Cr, Co explaining from 42% to 73% of the variation along with some association of Sb (22%), Cd (16%) and As (13%). These trace metals are typically emitted from a range of metal industries. The contribution of this factor was 1.3% ($0.11 \mu\text{g}/\text{m}^3$) to total measured HAPs on average. Substantial correlations of this factor were observed with Cu, Fe, Ba ($r = 0.47$ – 0.70) and notable correlations with EC, Zn, V, K⁺, Cl⁻ ($r = 0.20$ – 0.39) (Table 2). The CBPF plot (Fig. 2) revealed northeast and northwest as source directions under low wind speed ($<15 \text{ km/h}$) and westerly under high wind speed ($>20 \text{ km/h}$). Several metals processing operations located within 20 km to the west (e.g., Steel industries: Jackson, Westman), northwest (e.g., metal fabrication industries) and northeast (e.g., AltaSteel Ltd.) may be associated with this factor. There could also be minor contributions to this factor from the local Inland Concrete and Lehigh Cement plant based on a likelihood that Mn, Cr, Cd, As, Fe, Ba and Zn are emitted from this facility. All of these metals are listed as being emitted from Portland Cement kilns with electrostatic precipitators (which the local facility uses) (USEPA, 2016c). However, for the most part factor 5 was interpreted as a metals industry source.

3.2.6. Mixed urban

Factor 6 was distinguished by high levels of PAH species- BaA, Chr, BbF, BkF, BaP, DahA, IP, BghiP explaining from 42% to 70% of the variation and was attributed to 1.2% ($0.11 \mu\text{g}/\text{m}^3$) of the measured HAPs on average. PAHs are formed from incomplete combustion of organic matter such as oil, coal and wood and are released to the atmosphere from a variety of sources in urban areas including vehicle exhaust, wood-fired residential heating, fossil fuel industries and coke oven emissions (Harrison et al., 1996; Ravindra et al., 2008; Bari et al., 2009). This factor showed substantial correlations with NO₃⁻, EC, levoglucosan and mannosan ($r = 0.47$ – 0.56) and notable correlations with Ba, Cu, Zn, K⁺, and Cl⁻ ($r = 0.29$ – 0.40) (Table 2). The CBPF plot (Fig. 2) indicated south and northeast as notable directions under low wind speed ($<10 \text{ km/h}$) and northerly under high wind speed ($>20 \text{ km/h}$), suggesting a possible influence of local traffic, industries and wintertime residential wood combustion. Therefore, factor 6 was assigned as a mixed urban source.

3.3. Origin of air pollution event and non-event days

In this study, potential source origins of HAPs were further analyzed for 16 different event and non-event days during the 5-year study period. For this analysis, 24 h PM_{2.5} values $\geq 20 \mu\text{g}/\text{m}^3$ in the dataset were classified as 'event days', which is equivalent to exceeding a planning trigger for level 3 of the Canadian Ambient Air

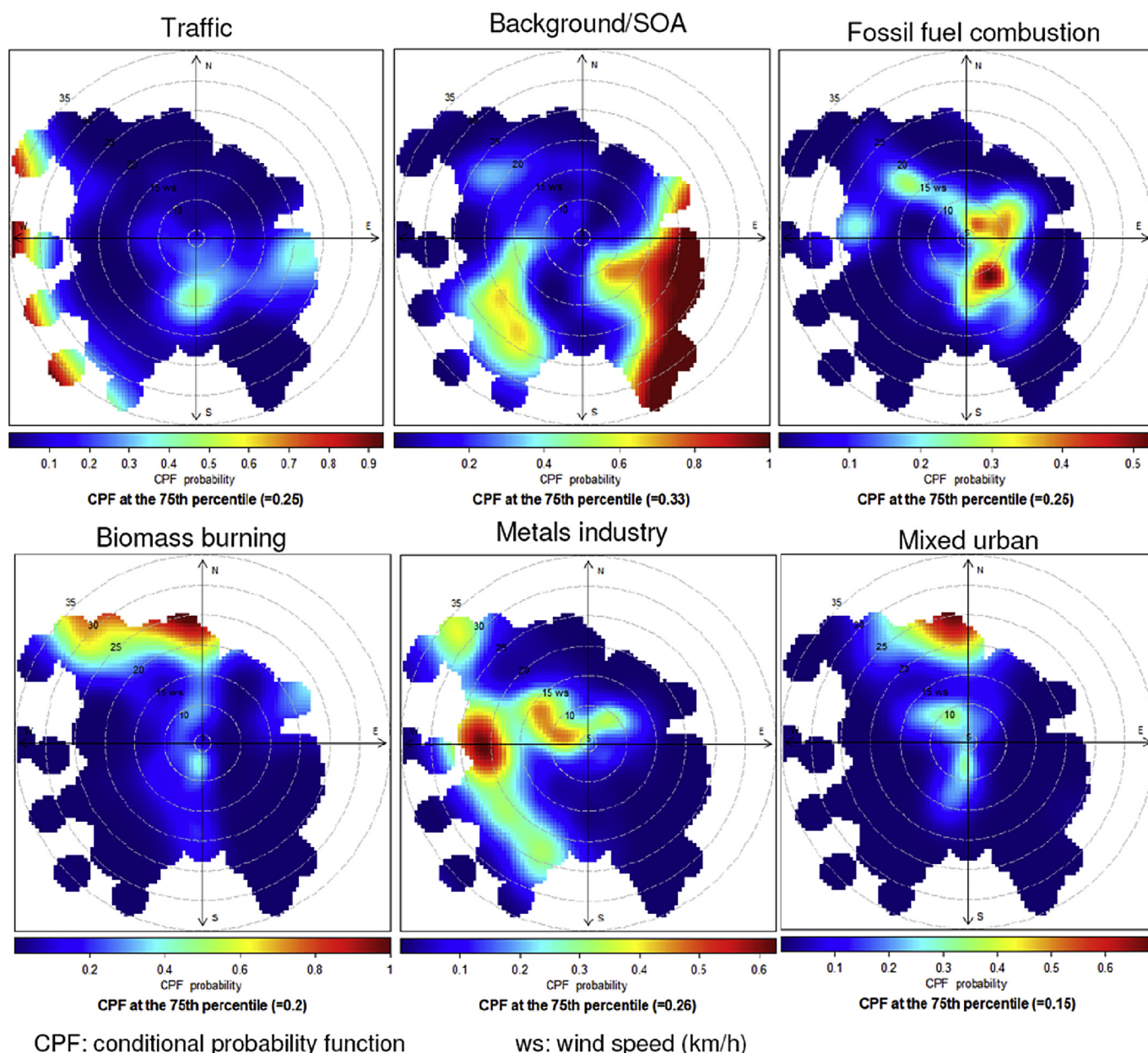


Fig. 2. CBPF plots for PMF-derived HAPs sources identified in Edmonton for 2009–2013.

Quality Standards (CAAQS) (CCME, 2012), and selected winter and summer 24 h $\text{PM}_{2.5}$ values $< 20 \mu\text{g}/\text{m}^3$ were classified as 'non-event' days. These daily events during winter and summer in Edmonton, the dominant PMF-derived HAPs sources and their potential source regions based on HYSPLIT modelling observation for the 24 h period are shown in Table 3 (Fig. 3, S13–S15).

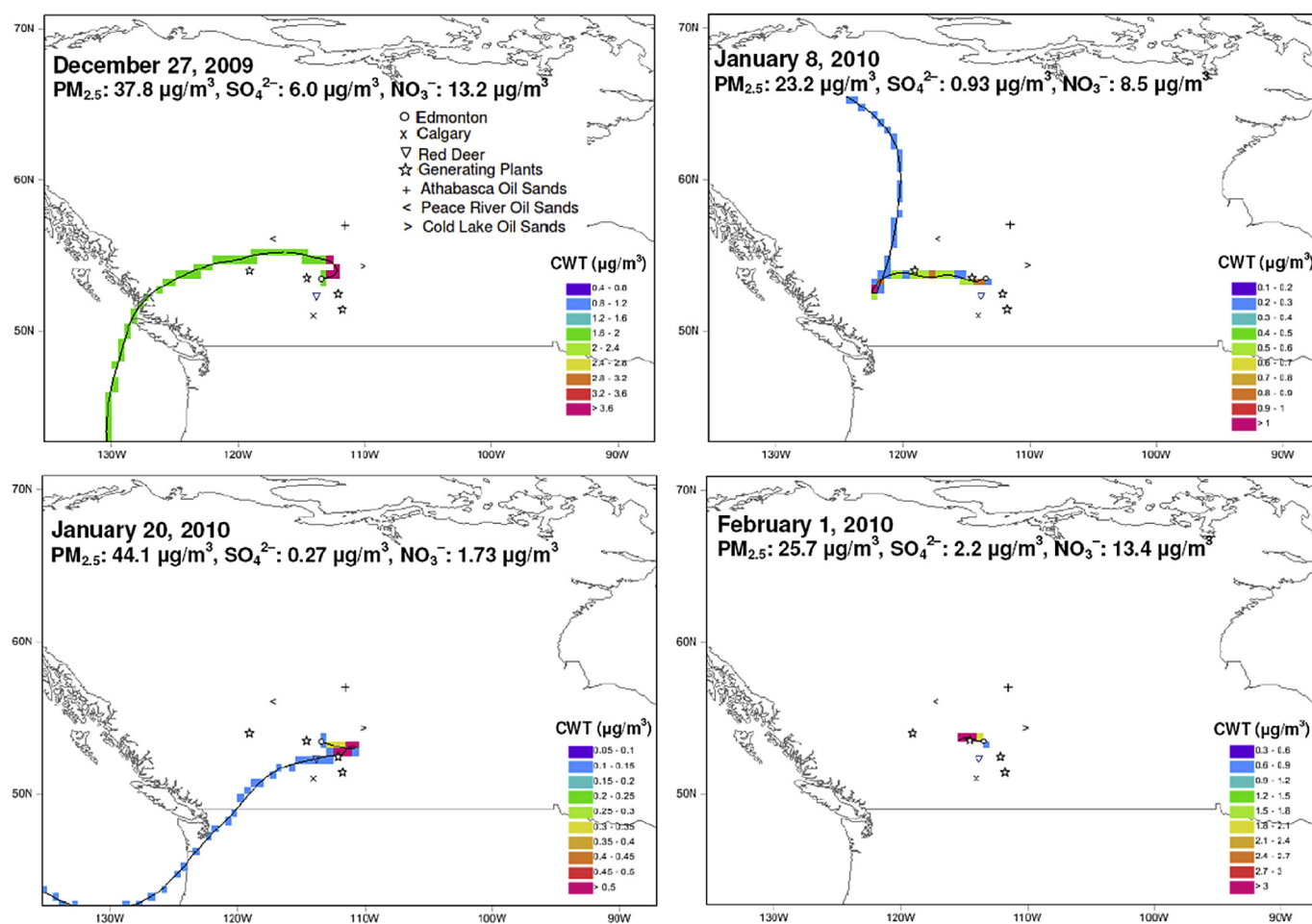
For the 5-year study period (2009–2013), traffic was identified as the most dominant contributor (42%) to $\text{PM}_{2.5}$ compared to background/SOA (25%) and fossil fuel combustion (11%) (Fig. 1). This suggests that local traffic plays a potentially important contribution to air toxics in Edmonton. For example, for event days listed in Table 3 with 24 h NO_3^- concentrations $> 4 \mu\text{g}/\text{m}^3$ (27-Dec-09, 8-Jan-10, 25-Feb-10, 3-Mar-10, 29-Oct-10, 22-Dec-10), local traffic was found as the dominant contributor to total HAPs compared to fossil fuel combustion and background/SOA. These findings are in contrast to a recent study undertaken in the Capital Region (AESRD, 2014) that reported $\text{PM}_{2.5}$ event-days at ambient air monitoring

stations in Edmonton were unrelated to traffic emissions from roadways near the monitors and the high concentrations may be due to secondary fine particulate matter from long-range sources.

Long-range transport of fossil fuel combustion also plays a role contributing to $\text{PM}_{2.5}$ event and non-event days and potential source regions were mostly immediately northeast of Edmonton, southern Alberta, northern Alberta (oil sands industry) as well as a minor influence from west of Edmonton (coal-fired generating plants). For the biomass burning source during summer, HYSPLIT-based CWT modelling identified the influence of forest smoke episodes from locations in southern Alberta on August 6, 2010 and British Columbia on August 24, 2010 (Fig. S13). It should be noted that due to the selection of available HAPs data including VOCs, PAHs, trace metals in the same days, some high $\text{PM}_{2.5}$ event days (e.g., 8-Jan-10: $62 \mu\text{g}/\text{m}^3$, 21-Aug-10: $62 \mu\text{g}/\text{m}^3$) were not included in the analysis, therefore their HYSPLIT trajectories were not presented here.

Table 3Dominant sources and potential source regions of HAPs on PM_{2.5} pollution event and non-event days in Edmonton for 2009–2013.

Date	PM _{2.5} ($\mu\text{g}/\text{m}^3$) ^a	SO ₄ ²⁻ ($\mu\text{g}/\text{m}^3$) ^a	NO ₃ ⁻ ($\mu\text{g}/\text{m}^3$) ^a	Dominant sources	Potential local and long-range source regions	Traffic $\mu\text{g}/\text{m}^3$	SOA $\mu\text{g}/\text{m}^3$	Fossil fuel combustion $\mu\text{g}/\text{m}^3$	Biomass burning $\mu\text{g}/\text{m}^3$	Evidence	SI
27-Dec-09	37.8	6.0	13.2	Fossil fuel combustion, traffic	Immediate northeast	12.59	0.20	9.48	1.43	HYSPLIT	Fig. 3
8-Jan-10	23.2	0.93	8.5	Traffic	Local	22.31	0.34	1.22	0.97	HYSPLIT	Fig. 3
20-Jan-10	44.1	0.27	1.7	Traffic	Local	11.79	2.35	0.74	0.80	HYSPLIT	Fig. 3
1-Feb-10	25.7	2.2	13.4	Fossil fuel combustion	West generating plants	6.86	1.64	4.47	−0.10	HYSPLIT	Fig. 3
25-Feb-10	25.2	0.86	10.6	Traffic	Local	13.04	2.34	0.99	0.35	HYSPLIT	Fig. S13
3-Mar-10	23.4	1.3	8.8	Traffic	Local	17.25	3.02	0.83	0.24	HYSPLIT	Fig. S13
6-Aug-10	22.5	1.7	0.08	Biomass burning, SOA	Southern Alberta forest fires	4.14	3.07	0.70	0.16	HYSPLIT	Fig. S13
24-Aug-10	7.1	0.26	n/a	Biomass burning, SOA	British Columbia forest fires	4.38	3.27	1.08	0.68	HYSPLIT	Fig. S13
29-Oct-10	23.0	6.6	4.1	Traffic	Local	8.82	2.39	0.96	0.43	HYSPLIT	Fig. S14
22-Dec-10	25.5	2.1	6.3	Traffic, fossil fuel combustion, biomass burning	Local, southern Alberta, U.S. regions	15.49	0.38	3.09	2.78	HYSPLIT	Fig. S14
10-Mar-11	18.9	4.5	6.6	Fossil fuel combustion	Northeast, south, southeast, southwest	1.08	1.33	2.38	−0.08	HYSPLIT	Fig. S14
9-Feb-12	20.0	4.4	3.6	Fossil fuel combustion	Athabasca and Cold Lake oil sands	3.12	1.43	1.32	0.56	HYSPLIT	Fig. S14
24-Sep-12	19.9	1.7	0.37	Traffic	Local	16.78	2.13	1.54	0.25	HYSPLIT	Fig. S15
17-Nov-12	25.5	1.0	12.8	Fossil fuel combustion, SOA	Immediate south, southwest, U.S. regions	2.14	2.84	2.34	0.26	HYSPLIT	Fig. S15
30-Nov-13	20.7	0.42	8.6	Biomass burning, fossil fuel combustion	Southern Alberta, British Columbia	3.63	1.64	1.85	3.08	HYSPLIT	Fig. S15
6-Dec-13	22.4	1.4	0.82	Biomass burning	Northern Alberta	0.87	0.09	0.53	6.66	HYSPLIT	Fig. S15

^a 24 h concentrations measured at Edmonton McIntyre station, n/a: not available, SI: supplemental information.**Fig. 3.** Potential source regions of fossil fuel combustion for PM_{2.5} event days.

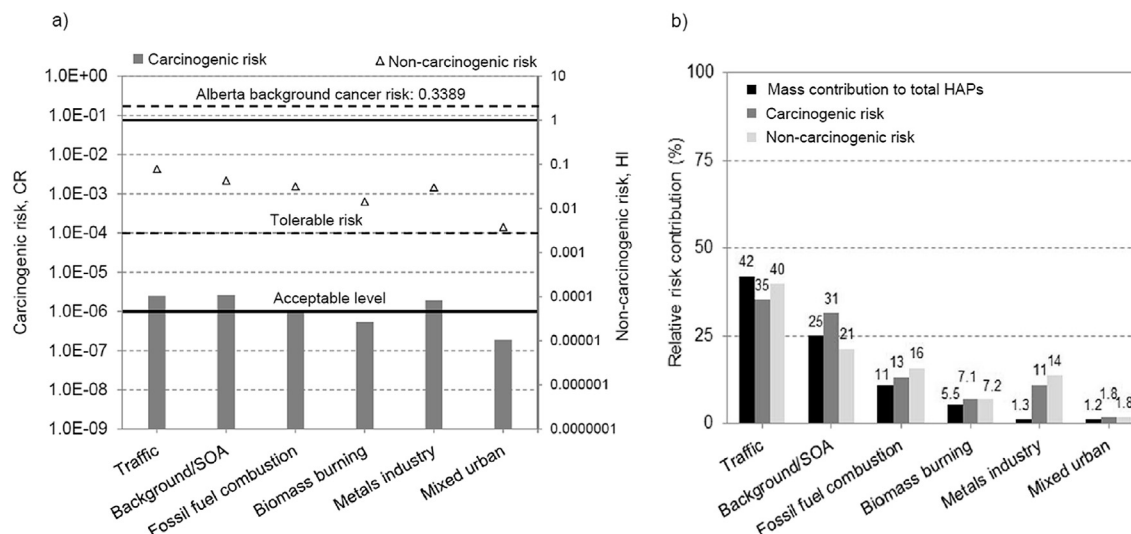


Fig. 4. Carcinogenic and non-carcinogenic risk estimates for identified HAPs sources and relative mass and risk contribution by each source.

3.4. Human health risk of HAPs sources

Health risk from inhalation exposure was apportioned to the identified six sources in Edmonton by summing carcinogenic and non-carcinogenic risks of all available risk-posing HAPs in a particular source and the source specific risk values are shown in Fig. 4a. Carcinogenic risk values of traffic, background/SOA and metals industry were above the USEPA acceptable level (1×10^{-6}), but below the tolerable risk (1×10^{-4}). Carcinogenic risks of other sources i.e., fossil fuel combustion, biomass burning and mixed urban were close to or below the acceptable level. Source specific non-carcinogenic risk values (HI) ranged from 0.004 to 0.08 and these were far below a safe level ($= 1$), suggesting that non-carcinogenic risks from HAPs sources are not expected to occur for people in Edmonton via inhalation.

The risk estimates by source contributions for HAPs were also assessed to determine the relative risk of identified sources in Edmonton. Fig. 4b shows the relative risk contributions of six identified sources in Edmonton over the study period. As reported previously (Fig. 1), traffic (42%) and background/SOA (25%) were the major contributors to total measured HAPs concentrations and they posed more risk to public health (66% and 61% of carcinogenic and non-carcinogenic risks, respectively). It is not surprising that fossil fuel combustion posed relatively low carcinogenic (13%) and non-carcinogenic (16%) risks consistent with previous risk estimates for PM_{2.5}-derived secondary sulfate and secondary nitrate sources (<10%) in Edmonton for the 2010–2015 period (Bari and Kindzierski, 2016).

Uncertainty in carcinogenic risk estimates of each source were also assessed using a bootstrapping technique after Wu et al. (2009) (Fig. S16). Mean risk estimates of all sources from the base PMF solution showed good alignment with the median of the distribution, suggesting less uncertainty in carcinogenic risk assessment. The 95th percentile of the risk estimates for fossil fuel combustion was just above the USEPA acceptable level (1×10^{-6}) but less than Alberta's incremental lifetime cancer risk (1×10^{-5}). These results suggest that potentially toxic HAPs in numerous sources identified in Edmonton were not a concern to public health during the 5-year study period (2009–2013). However, observed carcinogenic risk values of traffic and background/SOA were above the USEPA acceptable level.

We acknowledge that a limitation may exist in our risk apportionment approach. We only evaluated the health risk of HAPs sources based on available toxicity data for HAPs species and this may underestimate the total risk values. We investigated 17 available organic and trace metals (where 14 species were detected) in our study out of 30 most important air toxics (USEPA, 2016b) that pose the greatest potential threat to public health in urban areas.

4. Conclusion

To better understand current levels of air toxics, their sources and associated risk to public health, a study was carried out in the City of Edmonton over a 5-year period (2009–2013). Observed ambient concentrations of all air toxics were below the chronic and acute health risk screening criteria of the regulatory agencies and cancer benchmark targets in Alberta (1 in 100,000) except BaP, BbF and DahA. Carcinogenic risks of air toxics were also below the acceptable and/or within tolerable risks and non-carcinogenic risk were below the safe levels as recommended by regulatory agencies. Source apportionment analysis indicated that more than two-thirds of the total measured air toxics were attributed to local traffic (42%) and background/SOA (25%). Local traffic was found as a dominant contributor to high particulate air pollution event days in Edmonton particularly during winter months. Carcinogenic risk values for traffic, background/SOA and metals industry were above the USEPA acceptable level but below the Alberta benchmark suggesting that a considerable effort should be given to minimize the influence of traffic and industrial emissions.

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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.chemosphere.2016.12.157>.

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