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Sources Affecting VOC Levels in Quesnel, BC, Canada

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Abstract **-**In this paper, the sources contributing to the VOC concentrations in a small town in Quesnel, British Columbia, Canada are described. Quesnel is a small city with a population of 22000 inhabitants. The city sustained by forestry based industry. There are local plateaus around the downtown area which are at higher elevations than the downtown area that creates a bowl encompassing the downtown area. This topology of the town inhibits the air circulation in the valley. Volatile organic compounds (VOC) samples were collected on the roof of a secondary school located in the residential area in the city. 24-hour integrated ambient VOC samples were collected once every 3 days from June 2007 to February 2009. During this period, a total of 193 samples were collected. A receptor oriented model called Positive Matrix Factorization model was used to identify and quantify the sources in the region. Seven factors were identified. The highest contribution was due to space heating emissions. 22% of the modelled mass was attributed to this factor. The other identified factors were aged VOC, fossil fuel evaporation, industrial, motor vehicle exhaust, biogenic 1 and biogenic 2. At the end of the study, the location of the sampling equipment with respect to active stack at the roof of the secondary school was examined. It was determined that the major contribution for space heating emissions were due to emissions from the stack of the secondary school. This biased the obtained results. However, this study provided a valuable information how misplacement of the sampling location influence the source apportionment results.

Keywords**:** Source apportionment, Quesnel, Wood Based Industry, Volatile organic compounds

1. Introduction

Volatile organic compounds play an important role in the formation of secondary air pollutants such as ozone, peroxycarboxylic nitric anhydrides, aldehydes, and secondary organic aerosols in photochemical processes. Furthermore, many VOC species are hazardous air pollutants for human health. VOCs, together with nitrogen oxides (NOx) in the presence of sunlight, lead to the photochemical production of tropospheric ozone (Buzcu and Fraser, 2006). There are both anthropogenic and natural sources that may contribute to VOC concentrations in ambient air. Vegetation is the far most the important source of VOCs in the world. However, in cities, anthropogenic emissions dominates. The major anthropogenic emissions were from motor vehicles, industries and fossil fuel combustion for space heating.

Quesnel is the commercial centre of North Cariboo, located at the confluence of the Fraser and Quesnel rivers in British Columbia (BC), approximately 670 km north east of Vancouver. City and

metropolitan populations of Quesnel are approximately 10 000 and 22 000, respectively (BC Stats, 2012; Statistics Canada, 2007). The city is sustained by forestry based industries (primarily lumber, plywood, medium density fibreboard, and pulp), although mining and quarrying, ranching and tourism are important to the local economy (Quesnel Air Quality Roundtable, 2004). At the north and south ends of the valley, there are local plateaus which are at higher elevations than the downtown area that creates a bowl encompassing the downtown area (Quesnel Air Quality Roundtable, 2004). The local topography inhibits air circulation which can lead to frequent stagnation periods and build up of air pollutants in the valley (Plain, 1998).

Throughout the past two decades, Quesnel has attracted attention of researchers and local authorities due to high particulate matter (PM) levels in the region. The source apportionment of PM in the city was presented elsewhere (Doğan and Austin, 2012). The aim of this current study is to determine the sources of VOCs in the region using a receptor oriented source apportionment tool, namely Positive Matrix Factorization (PMF).

2. Experimental

2. 1. Sampling

Samples were collected on the roof of a secondary school located in a residential area in central Quesnel, BC (NAPS ID 101701). The sampling site was one of Canada's National Air Pollution Surveillance network sites and was operated by the British Columbia Ministry of Environment (Figure 1). The sampling station was located on the roof of Quesnel Secondary School (QSS), situated in a residential area. Cariboo Highway 97 and the railway more or less circle the air monitoring site. The highway passes approximately 470 m to the west, 820 m to the south, and 460 m to the east, whole the railway passes approximately 520 m to the east, 860 m to the south, and 415 m to the north. The commercial area of Quesnel is found south and west of QSS. Major industries (Cariboo Pulp, Quesnel River Pulp, Westpine MDF and Weldwood) were located approximately 1500 m northeast, 2200 m north, 3200 m northwest and 3700 m south of the sampling site, respectively.

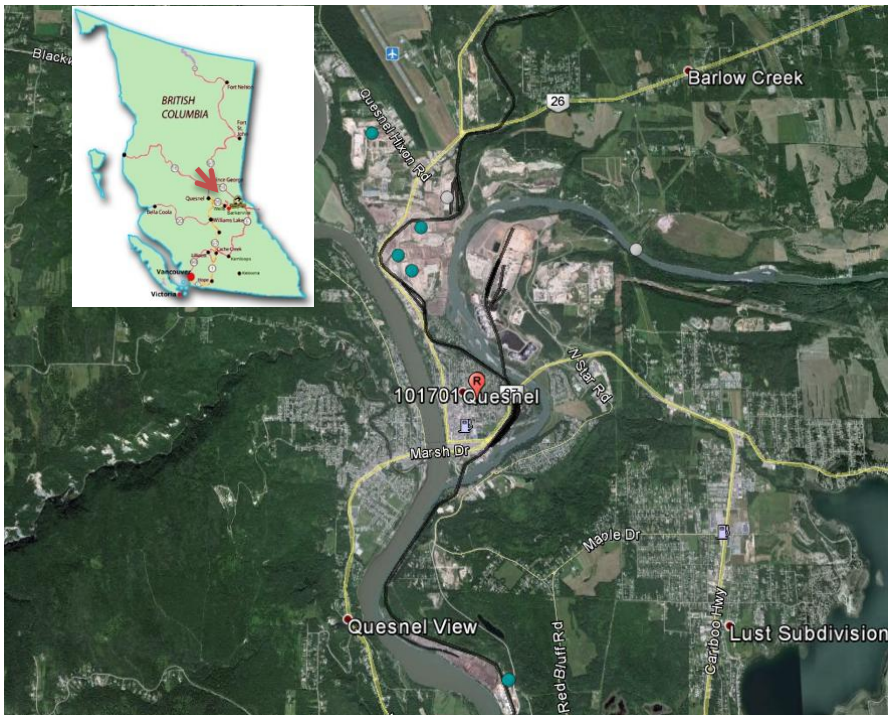


Fig. 1. Map of the air monitoring site, roadways (yellow), railways (black), and major industrial complexes located in Quesnel, BC, Canada (North Caribou Region, Caribou Regional District)

24-hour integrated ambient VOC samples were collected once every 3 days from June 2007 to February 2009. During this period, a total of 193 samples were collected in 6 L stainless steel SummaTM electro-polished canisters. A brief description of VOC sampling is given here, details on the sampling and analyses can be found elsewhere (Wang and Austin, 2006).

Samples were collected using whole air samplers (Xontech, Inc. Model 910A) and analyzed at the Analysis and Air Quality Division (Environmental Science & Technology Centre, Ottawa, Canada). Non-polar VOCs are stable in the Summa™ used in the NAPS network for collecting whole air samples. Polar VOCs are not stable in these canisters, but neither were they analysed. C3 to C12 hydrocarbons were analysed using automated cryogenic pre-concentration units (Nutech, Model 3550A, Nutech Corp., Research Triangle Park, NC or Entech Model 7100, Entech Corp., Calif.) and high resolution gas chromatography (Hewlett Packard 5890 Series II or Agilent 6890 GC) with quadrupole mass-selective detector (Hewlett Packard 5970 MSD or Agilent 5973) as described in EPA Method TO-14/15. The GC-MSD acquired data for target compounds only and ignored all others. Peak identification was based on GC retention time and MSD confirmation. Approximately 150 hydrocarbons were quantified in each sample.

2. 2. Source Apportionment

In this study, a receptor oriented method, Positive Matrix Factorization (PMF) model, was used to identify and quantify the sources. PMF is a multivariate factor analysis method (Paatero, 1997) that has been applied to a wide range of data, including 24-h speciated PM2.5, size-resolved aerosol data, deposition data, air toxics data, and VOCs (Kim and Hopke, 2004).

PMF uses a weighted least-squares fit with the known error estimates of the elements of the data matrix used to derive the weights. It produces quantitative non-negative solutions which can be written as:

$$X = GF + E \tag{1}$$

Or, by using the element-wise notation, bilinear factor analytic model is written as:

$$x_{ij} = \sum_{h=1}^{p} g_{ih} f_{hj} + e_{ij} \tag{2}$$

where X is known *i* x *j* matrix of the *j* measured chemical species in *i* samples. G is an *i x h* matrix of source contributions of the samples (time variation in source contribution). The F is an *h x j* matrix of the source compositions (source profiles). E is the residuals matrix, i.e., the difference between the measurement X and the model as a function of the factors G and F. PMF provides a solution that minimizes an object function, Q(E), based upon uncertainties for each observation. This function is defined as:

$$Q(E) = \sum_{i}^{n} \sum_{j}^{m} \left(\frac{e_{ij}}{s_{ij}} \right)^2 \tag{3}$$

where s_{ij} is an uncertainty estimate in the j^{th} element measured in the i^{th} sample.

For PMF modelling, two files are needed: uncertainty and data. For both of the files, detection limits should have to be known. However, detection limits for ethane, ethylene, acetylene, 2-methyl-1-pentene, 3-methyl-1-butene, 2-methyl-2-pentene, 2,5-dimethylheptane, 4-methyloctane, 3-methyloctane, 1-undecene, MTBE, a-pinene, b-pinene, d-limonene, camphene, Freon11 and bromochloromethane were not reported. The detection limits of these compounds were assumed to be 0.02 μg/m^3.

For a given compound, if the total number of BDLs and missing values was greater than 50% of all the observations, then that compound was not included in the analysis. For the remaining compounds, the

following procedure was applied. Measured values were directly used in the analysis. Depending on the concentration and chemical family (alkane, alkene, aromatic, halide), uncertainty was calculated by multiplying the concentration of the compound with the appropriate coefficient from Table 1 and adding the detection limit to the result (Paatero and Tapper, 1993). In order to not overestimate uncertainty, the mean uncertainty was calculated for each compound. Uncertainties of 1-pentene, 2-methyl-1-butene, 3-methyl-1-butene, cis-2-pentene, cyclopentene, cylcohexane, 2,4-dimethylhexane, 2,2,4-trimethyl pentane, nonane, isopropylbenzene, n-propylbenzene, 4-ethlytoluene, 1,3,5-trimethylbenzene, 2-ethyltoluene, 1,2,3-trimethylbenzene, indane, naphthalene, dodecane, freon114, chloroethane, chloroform, 1,2-dichloroethane, 1,1,1-trichloroethane and chlorobenzene were calculated without adding the detection limit to the results, because the measured values were close to detection limit. It would been unrealistic to add the DL to the uncertainty value which would have automatically increased the uncertainty by 100%.

Values below the nominal detection limit were replaced by half of the nominal detection limit values and their overall uncertainties were set to 5/6 of the nominal detection limit values. Missing values were replaced by the geometric mean of the measured values and their accompanying uncertainties were set at four times of this geometric mean value (Paatero and Tapper, 1993).

Table. 1. Uncertainty coefficients by chemical family and ambient concentration, with a 95% confidence level.

Concentration ($\mu g/m^3$)	Range	Alkane	Alkene	Aromatic	Halide
0.002-0.02		30	35	36	n/a
0.02-0.2		16	30	30	55
0.2-2		10	13	10	10
2 and up		11	10	9	7

2. 3. Supplementary Data

In order to identify sources of pollutants, supporting data are useful. For this purpose, concentrations of inorganic gaseous pollutants were used. Hourly data (NO, NO_2, SO_2, CO) were correlated with the source contribution (G score) data which was obtained from PMF analyses for the identification of sources.

3. Results and Discussion

After eliminating the compounds with 50% BDL, PMF was conducted for 81 compounds. Five to eight factor solutions were investigated. A seven factor solution was found to be the optimal solution for the Quesnel VOC dataset. In order to check model performance, observed mass was correlated with modelled mass. Quite high correlation between the modelled and observed mass was found (0.98).

The VOC sources were identified as space heating, aged VOCs, motor vehicle exhaust, gasoline evaporation, isoprene dominated biogenic emissions (Biogenic 1), a-pinene and b-pinene dominated biogenic emission (Biogenic 2), and industrial emissions (Table 2). The Pearson correlation coefficients between daily contributions of the factors with inorganic gases are given in Table 3.

The first factor was highly enriched with light VOCs, especially ethane, ethylene, acetylene, propane, 1,3-butadiene and butane, and benzene. Toluene and some halogenated VOCs were also present in this factor. Ethane is a well-known marker of natural gas (Song et al., 2008). Ethylene, acetylene and benzene are also major components from heating sources (Lau et al., 2010). This factor is highly correlated CO, NO and NO_2, indicating a combustion process. Therefore this factor is identified as space heating factor. This factor explains 22% (9.06 $\mu g/m^3$) of the total modelled mass.

The second factor was dominated by halogenated VOCs. Almost 60% of Freon22, chloromethane, Freon114, Freon113, chloroethane, Freon11, Freon12, dichloroethane, 111-trichloroethane, carbontetra chloride and chlorobenzene were explained in this factor. Ethane, undecane, naphthalene and dodecane also contributed to this factor. The halogenated VOCs have long lifetimes and are not reactive enough to

cause major changes in tropospheric ozone and secondary organic aerosol. Hence this factor is named as aged VOC. This factor contributed to 19% (7.75 μg/m^3) of the total VOCs.

Four- and 5-carbon VOCs were enriched in Factor 3, predominantly isobutane, butane, trans-2-butene, cis-2-butene, 2-methyl-1-butene, pentane, trans-2-pentene, cis-2-pentene, and 2-methyl-2-butene. These 4- and 5-carbon VOCs are indicators of evaporative emissions from fossil fuels. This factor is a result of gasoline evaporation and contributed to 10% (4.27 μg/m^3) of the VOC mass.

The fourth factor has a high loading of isoprene and low loadings of p-cymene, a-pinene, b-pinene, d-limonene, and camphene. Isoprene is a well-known marker of biogenic emissions and increases linearly with increasing photo-synthetically active radiation. Isoprene volatilization from vegetation appears to be controlled by temperature and light. Factor 4 explained 9% (3.88 μg/m^3) of the total VOC concentration.

Factor 5 explained most of the concentrations of a-pinene, b-pinene, d-limonene and p-cymene which are well known tracers of vegetation. Moderate loadings of camphene, indane and low levels of styrene were also observed in this factor. Although styrene is an indicator of industrial emissions, this factor was attributed to biogenic emissions of a,b-pinene. a-pinene and b-pinene emissions are related to temperature and relative humidity (Lamb et al., 1984). Lamb et al. (1984) reported that a-pinene emissions (primarily from Pseudotsuga menziesii) from wetted vegetation surfaces (rain or dew) were approximately 10 times larger than from dry branches at a given temperature. Fourteen percent (5.99 μg/m^3) of the VOC mass in the final dataset used for the PMF analysis was explained in this factor.

Factor 6 consisted mainly of aromatics and some alkanes, most of which are found in industrial solvents. Major contributors to this factor were 2,2,4-trimethylpentane,2-methylheptane, 3-methylheptane, 2,4-dimethylhexane, heptane, 3-methylhexane, 2,3-dimethylpentane, 2-methylhexane, methylcyclohexane, toluene, 2,2-dimethylbutane, and 2,4-dimethylpentane. Toluene and xylenes are especially well known markers of industrial emissions. Toluene concentration to benzene concentration in f-loading (T/B) for this factor was found as 7.7. Such high ratios indicates contribution of industrial emissions. Therefore, Factor 6 was attributed to mainly industrial activities in or near Quesnel. Factor 6 contributed 14% (5.72 μg/m^3) to total VOC mass.

Light VOCs, benzene, toluene, xylenes, ethylbenezene and some other aromatic VOCs were explained in Factor 7. Ethane, ethylene and BTEX compounds are motor vehicle exhaust markers. Light duty gasoline and heavy duty diesel vehicles differ in their emissions. In gasoline exhaust, acetylene, isobutene, isopentane, n-hexane, and 2-methylhexane are abundant whereas in diesel exhaust, propene, propane, 2,2-dimethylbutane, n-decane, and n-undecane are dominant (Watson et al. 2001). Both characteristics were observed in Factor 7. The toluene/ benzene ratio was checked in this factor and found to be 2.2. McLaren et al. (1996) reported a toluene/ benzene value for vehicle exhaust in Vancouver, BC, to be 1.8. These two values being comparable, this factor was attributed to motor vehicle emissions. This factor explained 12 % (5.18 μg/m^3) of the total modelled VOC.

3. 1. Effect of Monitoring Site Location on PMF Results

When selecting a monitoring site, many parameters must be considered, including, locality, terrain, meteorology, emission sources, possible chemical or physical interference, availability of services and site security. However, it is very difficult to locate an urban sampling site which satisfies all of these criteria. Although NAPS has guidelines for siting air monitoring stations for PM2.5 and ozone (Canadian Council of Ministers of the Environment, 2007), the guidelines are not specific enough to ensure that interferences will be avoided. Unfortunately, the siting of the air monitoring station in Quesnel resulted in biased VOC measurements. The sampling equipment was placed close to the roof access point, which was also within 9 m of the chimney stack from the furnace/boiler burning natural gas. This misplacement of the monitoring station leading to biased VOC measurements necessarily resulted in biased PMF results, also. The two most affected factors were space heating and fossil fuel evaporation factors.

Table. 2. Percent concentration of compound explained in each factor.

Compounds	Space Heating	Aged VOC	Gasoline Evaporation	Biogenic 1	Biogenic 2	Industrial Emissions
Ethane	48	35	1	0	2	0
Ethylene	47	2	0	1	1	22
Acetylene	52	7	0	1	1	18
Propylene	39	4	2	2	2	22
Propane	43	14	11	3	3	4
1-Propyne	52	0	0	0	0	18
Isobutane	34	9	39	4	0	0
1-Butene/Isobutene	31	5	24	3	1	12
1,3-Butadiene	46	0	3	0	1	18
Butane	33	8	39	6	0	8
trans-2-Butene	23	1	58	4	0	3
cis-2-Butene	21	3	56	5	0	8
Isopentane	19	6	41	9	0	22
1-Pentene	15	17	19	9	4	24
2-Methyl-1-butene	13	12	37	9	1	26
3-Methyl-1-butene	19	21	20	9	11	14
Pentane	27	8	31	8	1	19
Isoprene	5	2	0	87	0	0
trans-2-Pentene	10	5	41	11	2	26
cis-2-Pentene	11	8	38	10	1	27
2-Methyl-2-butene	11	3	42	9	3	25
2,2-Dimethylbutane	23	5	26	7	0	40
Cyclopentene	18	8	27	5	3	29
Cyclopentane	24	6	25	7	1	30
2,3-Dimethylbutane	22	5	26	8	0	38
2-Methylpentane	23	3	25	8	0	36
3-Methylpentane	22	4	22	7	0	37
Hexane	27	5	16	5	1	35
Methylcyclopentane	22	6	19	6	1	37
2,4-Dimethylpentane	21	2	13	6	1	40
Benzene	50	12	2	1	0	11
Cyclohexane	28	13	14	4	3	27
2-Methylhexane	20	3	10	6	1	42
2,3-Dimethylpentane	23	0	9	6	1	42
3-Methylhexane	20	2	9	6	1	43
2,2,4-Trimethylpentane	20	8	11	8	2	52
Heptane	23	2	8	6	3	45
Methylcyclohexane	25	9	8	8	4	41
2,4-Dimethylhexane	21	4	6	5	2	45
2,3,4-Trimethylpentane	24	14	4	6	3	35
Toluene	15	3	7	6	2	41
2-Methylheptane	21	2	6	6	1	47
3-Methylheptane	21	0	7	5	1	46
Octane	20	5	3	6	4	38
Ethylbenzene	10	3	7	4	1	36
m and p-Xylene	8	0	9	4	1	38
Styrene	0	12	0	10	22	22

Table 2 cont'd. Percent concentration of compound explained in each factor

Compounds	Space Heating	Aged VOC	Gasoline Evaporation	Biogenic 1	Biogenic 2	Industrial Emissions
o-Xylene	8	2	8	5	1	39
Nonane	13	4	7	5	3	26
iso-Propylbenzene	16	8	3	5	7	25
n-Propylbenzene	5	6	7	4	2	35
3-Ethyltoluene	3	2	8	4	4	38
4-Ethyltoluene	4	2	9	4	3	36
1,3,5-Trimethylbenzene	0	2	9	3	3	39
2-Ethyltoluene	4	2	8	4	2	37
1,2,4-Trimethylbenzene	0	4	9	4	3	36
Decane	7	15	7	6	2	11
1,2,3-Trimethylbenzene	0	5	6	10	14	25
p-Cymene	3	6	0	18	65	0
Indane	0	16	5	15	31	13
Undecane	2	23	4	5	1	0
Naphthalene	11	37	0	12	1	7
Dodecane	0	36	9	13	0	4
a-Pinene	0	4	2	15	72	2
b-Pinene	0	5	2	25	68	0
d-Limonene	0	2	1	14	80	2
Camphene	4	18	0	17	39	8
Freon22	21	57	0	8	4	7
Chloromethane	21	64	1	8	4	2
Freon114	21	63	1	7	4	4
Freon113	21	63	1	7	4	4
Chloroethane	17	66	0	8	1	0
Freon11	21	62	1	7	4	3
Freon12	19	63	1	8	4	5
Dichloromethane	20	59	0	6	1	4
Chloroform	19	53	1	7	7	7
1,2-Dichloroethane	23	56	2	4	3	0
1,1,1-Trichloroethane	15	61	2	7	4	9
Carbontetrachloride	17	63	2	7	5	5
Tetrachloroethylene	25	0	11	0	2	12
Chlorobenzene	14	63	0	5	4	0

Table. 3. Pearson correlation coefficients (R) of VOC source contributions with inorganic gases.

Sources	CO	NO	NO_2	SO_2
Factor 1- Space Heating	0.72^{**}	0.81^{**}	0.85^{**}	0.15^{**}
Factor 2- Aged VOC	-0.64^{**}	-0.64^{**}	-0.65^{**}	-0.46^{**}
Factor 3- Fossil Fuel Evaporation	0.53^{**}	0.57^{**}	0.48^{**}	0.20
Factor 4-Biogenic 1	-0.26	-0.23^{**}	-0.20^{**}	0.39^{**}
Factor 5- Biogenic 2	-0.25^{**}	-0.19^{*}	-0.22^{**}	0.29^{**}
Factor 6- Industrial	0.59^{**}	0.63^{**}	0.55^{**}	0.11
Factor 7- Motor Vehicle Exhaust	0.73^{**}	0.68^{**}	0.63^{**}	0.12

**Correlation is significant at the 0.01 level *Correlation is significant at the 0.05 level

4. Conclusion

In this study, PMF was applied to both ambient VOC data sampled between 2007 and 2009 in Quesnel. Seven factors were found to influence the ambient VOC concentrations of Quesnel. Space heating by natural gas (22%) was also found to be the predominant VOC source in Quesnel. Other sources included aged VOC (19%), fossil fuel evaporation (10%), two biogenic factors (23%), industrial (14%) and motor vehicle emissions (12%). Two biogenic factors were found in the area and the differences between the emission trends were documented. Aged VOC contribution is found to be higher than expected. This could be because of lower emissions from other sources. The industrial factor was not attributed to any specific industry as the indicators were not significant in the source profile. Sampling location biased the contributions of gasoline evaporation and space heating factors.

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