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Contribution to $PM_{2.5}$ from domestic wood burning in a small community in Sweden

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Abstract

Biomass burning for domestic heating has increased in many countries with cold climates in recent years. This paper presents and compares two ways of estimating the contribution of particulate matter ($PM_{2.5}$) to ambient air from local domestic wood burning, using daily stationary parallel $PM_{2.5}$ measurements in a wood-burning area and at a reference location. In the first method (based on air mass back trajectories), daily gravimetric $PM_{2.5}$ mass differences were compared between the two stations for days with low contribution from regional sources. In the second method, 28 filters from each location were chemically analysed, and source contributions were calculated using positive matrix factorisation (PMF). The trajectory method estimated the extra local contribution from domestic wood burning in the wood-burning area to be 0.7–1.1 µg m^{-3}, while the PMF method gave a contribution of 0.64 µg m^{-3}. With the PMF method, the total contribution to ambient air from local domestic wood burning was estimated to be 25% of the total $PM_{2.5}$ mass. The estimated mass contribution using the trajectory method gave a result similar to that of the PMF method, and the method can therefore be a time- and cost-effective first step, especially when no chemical analysis is possible.

1. Introduction

The use of wood-burning appliances (boilers, stoves, and fireplaces) for heating is quite common in the homes of the countries with cold climate. Biomass burning as the main or supplementary domestic heating source in colder seasons has increased over the past decade in the Nordic countries. The increasing cost of both fossil fuels and electricity has been an important factor driving this increased biomass burning. Moreover, it has been government policy in several countries to promote a shift to alternate fuel consumption as a measure to decrease dependency on fossil fuels and on unsustainably produced electricity. The increase of biomass burning for space heating has made it one of the major sources of fine particulate matter ($PM_{2.5}$), and the Clean Air for Europe (CAFE) programme[1] has estimated that in the year 2000 25% of the primary $PM_{2.5}$ emissions in the EU countries came from domestic wood burning. Based on this figure, it is estimated that in 2020, 38% will come from domestic wood burning, making it the largest source of $PM_{2.5}$.

Biomass burning produces a number of gaseous and particulate pollutants which affect the ambient air.[2-5] Emissions from wood burning vary considerably, depending on wood type, combustion appliance, combustion conditions, and phase of the combustion cycle.[5-7] The pollutants are emitted to both the indoor and ambient air, and can affect both the owners and their neighbours.[8-10] The relative toxicity of PM from combustion of wood compared to other PM is unclear. There is limited evidence as to which emission characteristics are important for health effects. Two separate review articles on the subject[7, 11] have concluded that PM from wood smoke seems to be at least as harmful as PM derived from other sources.

The rising importance of biomass emissions in ambient air increases the need to quantify the contribution in local residential areas. Simple methods for such quantifications would facilitate the scientific understanding of the contribution from biomass PM.

The aims of the present study were to investigate and quantify the contribution to ambient air from domestic biomass burning in a residential area with a history of complaints, and to compare the effectiveness of a simple quantification method (using mass concentration data and trajectory analysis) to a more advanced source apportionment method (positive matrix factorisation, PMF).

2. Material and methods

2.1. Study design

The study took place in Tanumshede, a small community with 5000 inhabitants, 135 km north of Gothenburg and 6 km from the Swedish west coast (N 58° 43', E 11° 20') during the winter season, 17 November 2007 to 15 April 2008. Daily sampling of $PM_{2.5}$ was performed at two fixed outdoor stations, in a wood-burning area and at a reference station. The wood-burning station was located in the middle of a district with 80 single-family houses where domestic burning for heating was common, and the reference station was located in the outskirts of the community, about 2 km southeast of the wood-burning station, with no domestic wood burning in the vicinity. The station in the wood-burning area was chosen because of the high density of domestic wood burning and due to unfavourable topographic conditions in this area. A map of the Swedish west coast and an aerial photograph covering the sampling sites are available in the supplementary file.

In the wood-burning area, 35% of the homes had some type of biomass burning appliance for heating (main or supplementary), and 72% of the homes had at least one neighbour within 50 m; an additional 20% had at least one neighbour within 200 m with a biomass-burning appliance.

2.2. Monitoring

Daily 24-hour $PM_{2.5}$ samples were obtained using the IVL weekly $PM_{2.5}$ sampler (IVL Swedish Environmental Research Institute, Gothenburg, Sweden) on 25 mm TF (PTFE) membrane filters (Pall Corp., Port Washington, NY, USA) at the two sampling stations. Out of a total of 151 days, matched filter samples were successfully collected for 135 days. At the wood-burning station, temperature and relative humidity were measured continuously with a resolution of 1 hour at two heights, 3 and 10 m above ground, using Tinytag Plus 2 TGP-4500, dual-channel temperature/relative humidity logger (Gemini Data Loggers, Chichester, West Sussex, UK). From these data it is possible to identify temperature inversion events.

2.3. Meteorology

During the sampling period only 30 days (out of 151) had a mean temperature below freezing, and only one day had fully snow-covered ground this winter, compared to the 50–75 days common in this part of Sweden (mean over the years 1961–1990).[11] The mean temperatures in Gothenburg for December, January, and February, were 3–5°C warmer than usual (Gothenburg environmental office). These features affect both the emissions and the dispersion of the emitted wood smoke.

2.4. Air mass back trajectories

Air mass back trajectories were computed using the NOAA ARL HYSPLIT model[12] to investigate the effect of long-range transported pollution. For each sampling day, 96-hour air mass back trajectories were computed at start-up time, and at 12 and 24 hours later. The trajectories were divided into four classes representing different source areas, namely, continental areas (Central and Eastern Europe); marine areas; the Nordic countries Finland, Norway, and Sweden; and the UK, or remained undetermined (for trajectories that shifted classes during the sampling day) as previously described.[13] The classification was made according to the criterion that all trajectories during a sampling period must have a major path belonging to the same class.

2.5. Analytical techniques

The filters were weighed at the IVL lab before and after sampling. The filters were conditioned for 24 hours at a temperature of 19–21°C and relative humidity of 47–53%. After mass concentration was determined, a subset of the filters (56 filters paired from 28 sampling days) was selected for analysis of black carbon (BC) and elemental concentrations. The subset was chosen among days identified as being characterised by Nordic or marine air masses (low levels of air pollution). This selection was made in order to minimise the contribution (and noise) from long-range transported polluted air.

Analysis of BC was made using a Model OT21 optical transmissometer (Magee Scientific Corp., Berkeley, CA, USA). An energy-dispersive X-ray fluorescence (EDXRF) spectrometer at the Department of Chemistry, Atmospheric Science, University of Gothenburg, was used to analyse the elemental composition of all filter samples.[10] The EDXRF spectra were processed and quantified using the quantitative X-ray analysis system (QXAS) and the analysis of X-ray spectra by iterative least-square fitting (AXIL).[14] All samples were analysed using a live time of 1000 seconds, a tube voltage of 55 kV, a tube current of 25 mA, and a Mo secondary target.

2.6. PMF source apportionment

Source apportionment analysis was carried out by positive matrix factorisation (PMF) technique using the US Environmental Protection Agency software EPA-PMF 3.0.[15] PMF is a multivariate receptor model concept that estimates the source profiles and their contributions based on a weighted least-square approach.[16, 17] The following are the basic equations of the PMF model, which have been developed to obtain the unknown matrices, G and F, by the iterative treatment of a least-square method:

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

$$Q(E) = \sum_{i=1}^{m} \sum_{j=1}^{n} \left(\frac{e_{ij}}{s_{ij}} \right)^2 \tag{2}$$

X is the data matrix (size m × n) consisting of m chemical components analysed in n samples, G is the source contribution to each sample (size n × p) for p factors, and F is the matrix of the source profile (size p × m). The matrix E is the residual. The main task of the PMF is to minimise the Q-value, which is defined in the equation (2) as the sum of the square of the residuals (e_{ij}) weighted inversely with the error estimates (s_{ij}) of the data point.

In the present study the following chemical components were used: BC, S, Cl, K, Ca, Ti, Mn, Fe, Ni, Cu, Zn, Br, and Pb. For the PMF models, the elemental concentrations have been recalculated to their mean oxidised concentrations when applicable.[18] The following settings were used when running the PMF. If the signal-to-noise ratio was below 2 for a variable in the model, the variable was deemed weak (and the uncertainty was tripled). $PM_{2.5}$ mass was specified as a ‘total variable’, that is, the mass concentrations of the other variables were subtracted from the $PM_{2.5}$ mass. The uncertainty for the $PM_{2.5}$ mass was automatically tripled, because a total variable should not have a large influence on the solution.

2.1. Statistical analysis

Statistical calculations were performed with the SAS system for Windows, version 9.2.[19] Correlations between elemental concentrations in different microenvironments were assessed using the Spearman rank correlation coefficient (r_s), and differences between pairs of samples using the Wilcoxon signed rank test. For unpaired observations, Wilcoxon's rank sum test was used. Statistical significance refers to $p < 0.05$ in two-tailed tests. If the elemental concentration was below the limit of detection (LoD), the LoD divided by the square root of two was used in the calculations.[20]

3. Results

3.1. $PM_{2.5}$ mass concentration and trajectory analysis

The levels of $PM_{2.5}$ in this small community were low. Mean concentration during the winter was 5.6 $\mu g\ m^{-3}$ in the wood-burning area (range 1.3–20 $\mu g\ m^{-3}$), and similar for the reference station (Figure 1). The results of $PM_{2.5}$ mass concentrations at the two stations for the full data set, and for the division into trajectory classes, cold/warm days, and days with positive/negative temperature profiles are presented in Table 1.

For days when the air masses coming to Tanumshede originated from continental Europe, the mean concentration increased to 7.9 $\mu g\ m^{-3}$ (a 40% increase), while if the air originated from the north Atlantic (marine air masses), the levels were lower (4.7 $\mu g\ m^{-3}$). On cold days compared to warm, a mean difference of 0.6 $\mu g\ m^{-3}$ was found. However, on days with a positive temperature gradient (i.e. a potential for ground level inversion) only a minor increase was found.

No statistical difference was found for the whole winter season (N = 135 days) between the wood-burning area and the reference location; however, when the time series was divided into subcategories according to air mass origin, temperature, or possible inversion, several statistically significant differences were found. On days with air masses of Nordic or marine origin (the trajectory method), higher $PM_{2.5}$ concentrations, 0.7 and 1.1 $\mu g\ m^{-3}$, respectively, were found in the wood-burning area (Table 1). When the daily mean temperature was <0°C, a significant mean increase of $PM_{2.5}$ levels of 0.9 $\mu g\ m^{-3}$ was noted, and days with a positive temperature profile had a small but still significant increase (Table 1).

3.2. Elemental analysis

For the subset of the filter samples, BC and elemental concentrations were determined to investigate whether there were any significant differences for any elements related to biomass burning. The BC and elemental concentrations as well as $PM_{2.5}$ mass for this subset of the data are presented in Table 2.

For this subset of paired samples, statistical analyses were performed on the whole subset (Table 2), and also divided by trajectory class, temperature, and positive temperature profile (the results are available in Table S1 in the supplementary file). Significantly higher concentrations were found in the wood-burning area compared to the reference station in the whole subset for $PM_{2.5}$, BC, K, Zn, and Br (Table 2). In addition, higher concentrations were also found for $PM_{2.5}$ on days with Nordic air masses, and on days with a positive temperature profile, and also for BC, K, and Zn for all comparisons of the subsets (see Table S1 in the supplementary file).

3.3. Positive matrix factorisation

The PMF analysis on the combined dataset from both the wood-burning area and the reference location (n = 56) resulted in a model with five sources affecting the sampled $PM_{2.5}$ mass. The sources were, in order of mean contribution: long-range transported (LRT), biomass burning, sea salt, soil, and traffic. Profiles of the sources and the species contribution to each profile are presented in Figure 2.

The LRT particles had high concentrations of S, Pb, Br, and Ni. LRT made up on average 38% of the total mass contribution. Biomass burning contributed 26% and had high concentrations of BC, K, and Zn. Sea salt is dominated by Cl, and the contribution was 21%. The remaining factors, soil and traffic, had minor impact on the sampled mass, and their contributions were 12% and 4%, respectively. Altogether the five-factor model accounted for 84% of the total $PM_{2.5}$ mass.

By separating the model output from the subset of the analysed filters into those from the wood-burning area and those from the reference location (the PMF method), one can compare how the different sources impacted the two areas (Figure 3).

The main difference between the two locations is the contribution from biomass burning. In the wood-burning area the contribution was 1.41 $\mu g\ m^{-3}$ compared to 0.77 $\mu g\ m^{-3}$ in the reference location. It is evident that the contributions from all factors besides biomass burning were very similar and that the excess contribution from biomass burning, 0.64 $\mu g\ m^{-3}$, in the wood-burning area explained the difference in $PM_{2.5}$ mass.

4. Discussion

The simpler method of $PM_{2.5}$ mass analysis on data subsets classified by the trajectory computations (trajectory method) gave an estimate of the local contribution in the wood-burning area similar to that of the more costly and laborious method of chemical analysis of $PM_{2.5}$ followed by PMF modelling (PMF method), 0.7–1.1 $\mu g\ m^{-3}$ vs. 0.64 $\mu g\ m^{-3}$, respectively. It has been estimated by Forsberg et al.[21] that 50%–75% of PM_{10} in Sweden originates from abroad, and mainly from continental Europe. The trajectory method can therefore be used to identify days when the local sources play a more important role due to the long-range transported contribution being lower. These facts makes it possible to use existing monitoring data of $PM_{2.5}$ to estimate the contribution from local biomass burning without the need to start new sampling programmes and use costly analytical methods, at least, as a first step, provided that suitable monitoring stations are available. This could be beneficial to both local municipal environmental offices and regional and national monitoring networks. A second option, if monitoring data using a filter-based technique is available, could be to select, by trajectory analysis, a smaller subset of filters to analyse the chemical characteristics for later PMF source apportionment.

4.1. Markers for wood smoke

Consistently higher concentrations were found in the wood-burning area for BC, and for the elements K and Zn. In several studies these compounds have been shown to be good markers for biomass burning.[3, 10, 22, 23] The median differences between the two areas for these compounds (see Table 2) are similar to those found indoors in an earlier study[10] suggesting that the contribution from domestic wood-burning is realistic in this type of settling.

The $PM_{2.5}$ levels were low, but comparable to what can be found in Gothenburg on days with similar conditions.[13] The levels varied, depending on the source contribution from regional transport. When the air masses originated from more densely populated and industrialised areas (i.e. central Europe or the UK), higher $PM_{2.5}$ levels were found (Table 1). On those days there was an increase of combustion-type aerosols that can mask the local wood combustion signal. In order to minimise the influence from regional combustion sources, only days with low impact of regional transport (Nordic and marine air) were chosen for the chemical analysis. The $PM_{2.5}$ levels for those types of air masses (median levels 5.0 and 4.6 µg m^{-3} for Nordic and marine) were almost the same as those found in a study in Gothenburg a few years earlier, median ambient $PM_{2.5}$ level of 5.1 and 4.9 µg m^{-3} for Nordic and marine air, respectively.[24]

4.2. Wood smoke contribution

The PMF analysis of the data resulted in five source factors, LRT, biomass burning, sea salt, soil, and traffic, listed from highest to lowest. All factors, besides the biomass burning, were very similar in source strength at both sites, indicating that the only real difference between the two sites was the presence of biomass burning in the wood-burning area. The reference area was not devoid of any biomass influence, but the contribution was only about half compared to the wood-burning area. The absolute contribution from domestic wood-burning of 1.4 µg m^{-3} was slightly smaller compared to what has been presented in other studies in the Scandinavian countries. In the small community of Hagfors in central Sweden a 2 µg m^{-3} increase in a wood-burning area was found compared to a district heating area.[25] In the town of Lycksele in northern Sweden the estimated contribution from residential biomass was about 3 µg m^{-3}.[26] In a study conducted in a small community in Denmark,[27] the local wood-smoke contribution was estimated to be 4 µg m^{-3}, when comparing the levels with a nearby background station. The most likely reason for the somewhat lower levels in the present study is the mild winter season in 2007–2008. The unusually warm winter affected the total amount of biomass burnt, and the rain and wind scavenged and dispersed the emitted smoke more efficiently. Nevertheless, we found a statistically significant contribution from domestic biomass burning to the $PM_{2.5}$ mass under several different conditions. Using the PMF method, the average contribution from biomass burning in the wood-burning area was found to be 25% of the total $PM_{2.5}$ mass (1.4 µg m^{-3} vs. the season average of 5.6 µg m^{-3}), and the highest daily contribution was 66% (data not shown); thus biomass burning had a substantial influence on the $PM_{2.5}$ levels. Glasius et al.[27] also found an average mass contribution of about 25% in their study, but a much higher contribution (82%) was found in Libby, Montana.[28] In a review by Kochbach Bolling et al. and references therein it is stated that biomass combustion contributes 10–40% of $PM_{2.5}$ in larger cities, but the daily contribution can be much higher (80–90%).[6]

The contribution from LRT in the current PMF model was smaller than in reality for the winter season, since the analysis was performed only on a subset of the data without any influence from the more polluted regions of Europe. For all the other sources, including domestic biomass burning, the contribution is believed to be representative for the winter season, since they were mostly of local or nearby origin.

5. Conclusions

The average contribution to local $PM_{2.5}$ levels from domestic biomass burning for space heating was found to be 1.4 µg m^{-3}, and this was 0.64 µg m^{-3} higher than at the reference location. By using air mass back trajectory analysis and selecting days with lower levels of regional transport of pollutants, statistically significant differences were found. The difference in $PM_{2.5}$ mass concentration between the two locations for days with air masses from Nordic or marine origin was 0.7 and 1.1 µg m^{-3}, respectively. This simpler method gave results similar to those of PMF modelling and can be an alternative when monitoring data are available but no chemical analysis is possible (e.g. by local municipal monitoring programmes).

6. Acknowledgement

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Tables

Table 1. Mean $PM_{2.5}$ concentrations (µg m^{-3}) for the whole data set and for the different subsets. Median concentrations are within parentheses, and mean differences (wood-burning minus reference) marked in bold were statistically significant ($p<0.05$ Wilcoxon signed rank test).

	# Days	Wood-burning	Reference	Difference
All data	135	5.6 (4.9)	5.5 (4.5)	0.1
By trajectory class				
Nordic	27	5.7 (5.0)	5.0 (4.5)	**0.7**
Marine	30	4.7 (4.6)	3.6 (3.1)	**1.1**
Continental	28	7.9 (6.4)	7.6 (6.7)	0.2
UK	26	5.2 (4.7)	6 (4.9)	-0.9
Undetermined	24	4.7 (3.7)	5.7 (5.1)	-1.0
By temperature				
Cold (<0°C)	28	6.1 (5.4)	5.3 (4.3)	**0.9**
Warm (>0°C)	107	5.5 (4.9)	5.6 (4.6)	-0.1
By temperature gradient				
Inversion	87	5.6 (4.9)	5.3 (4.2)	**0.3**
No Inversion	48	5.8 (5.0)	6.0 (5.3)	-0.2

Table 2. Median concentrations and ranges (in µg m^{-3}) for $PM_{2.5}$, BC, and the elements in the wood-burning area (N = 28) and at the reference station (N = 28). Median differences of pairs marked in bold were statistically significant ($p < 0.05$ Wilcoxon signed rank test).

	Wood-burning area		Reference station		Median difference of pairs
	Median	Range	Median	Range	
$PM_{2.5}$	4.9	(1.8–11.4)	3.9	(1.8–12.4)	**1.3**
BC	0.34	(0.03–0.92)	0.21	(0.02–0.57)	**0.078**
S	0.70	(0.30–1.87)	0.64	(0.3–2.32)	0.072
Cl	0.09	(0.06–3.09)	0.10	(0.07–3.53)	-0.002
K	0.22	(0.07–0.65)	0.17	(0.05–0.57)	**0.10**
Ca	0.095	(0.04–0.26)	0.094	(0.02–0.29)	0.001
Ti	0.025	(0.014–0.064)	0.022	(0.014–0.049)	0
Mn	0.0053	(0–0.013)	0.0042	(0–0.014)	0
Fe	0.059	(0.013–0.282)	0.049	(0.007–0.265)	0.005
Ni	0.0027	(0.0001–0.006)	0.0017	(0.0002–0.005)	0
Cu	0.0031	(0.0001–0.0153)	0.0042	(0.0009–0.0219)	-0.001
Zn	0.034	(0.007–0.11)	0.025	(0.004–0.085)	**0.009**
Br	0.0027	(0.0013–0.0082)	0.0018	(0.0009–0.0062)	**0.001**
Pb	0.0058	(0.0005–0.023)	0.0052	(0.0012–0.012)	0.001

Figures

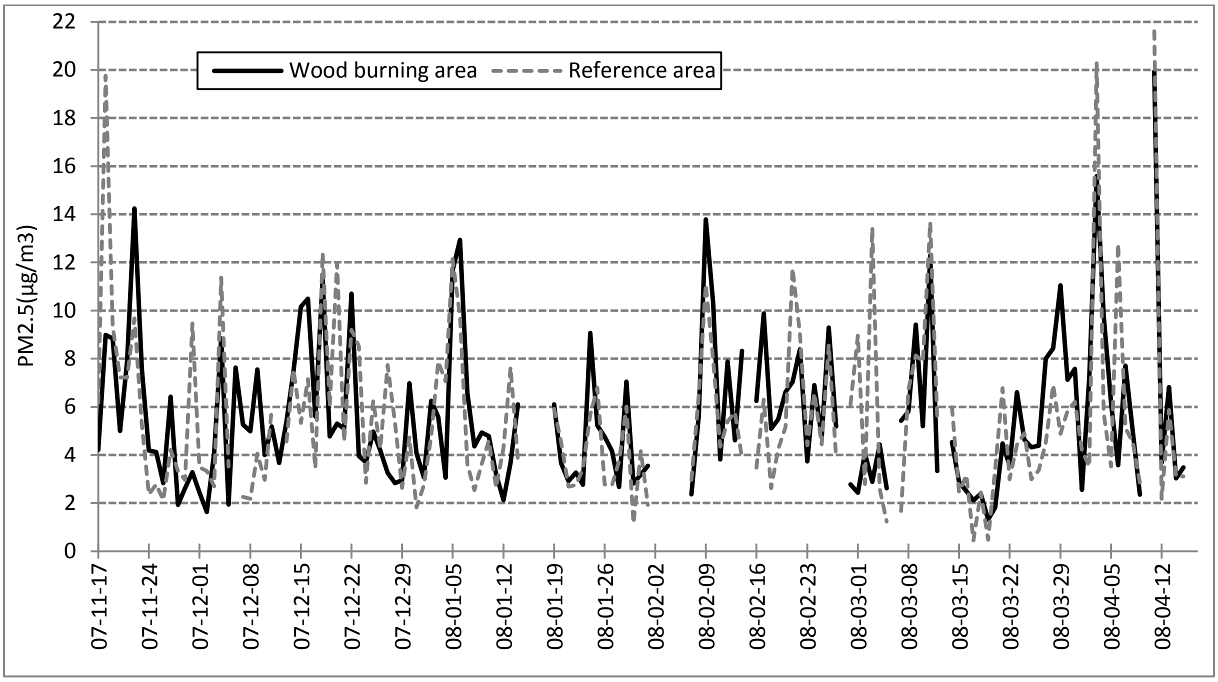

Figure 1. Time series for the wood-burning area and the reference station.

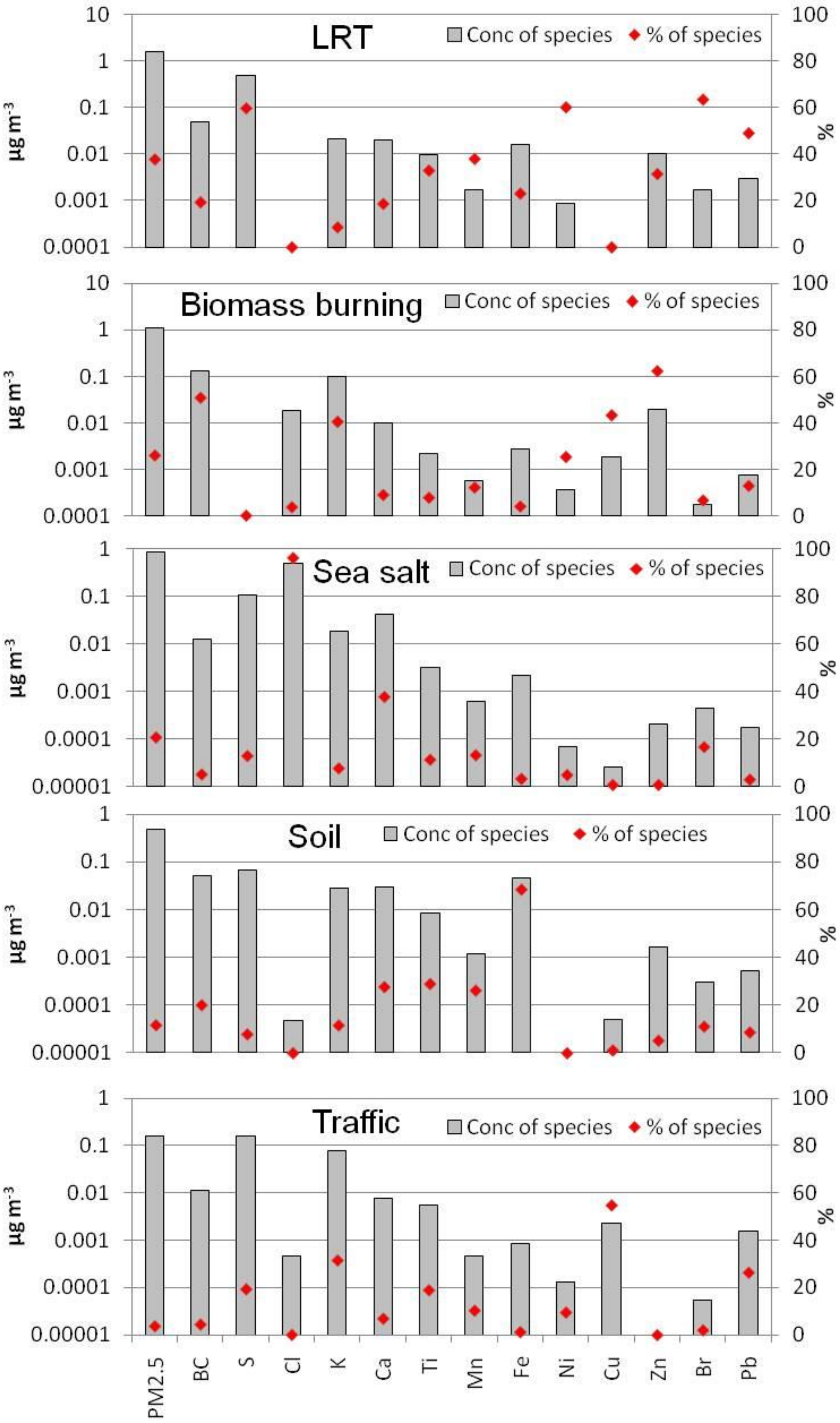

Figure 2. Profiles and contributions of the species in each source from the combined five-factor PMF model on the 28 + 28 paired filter samples.

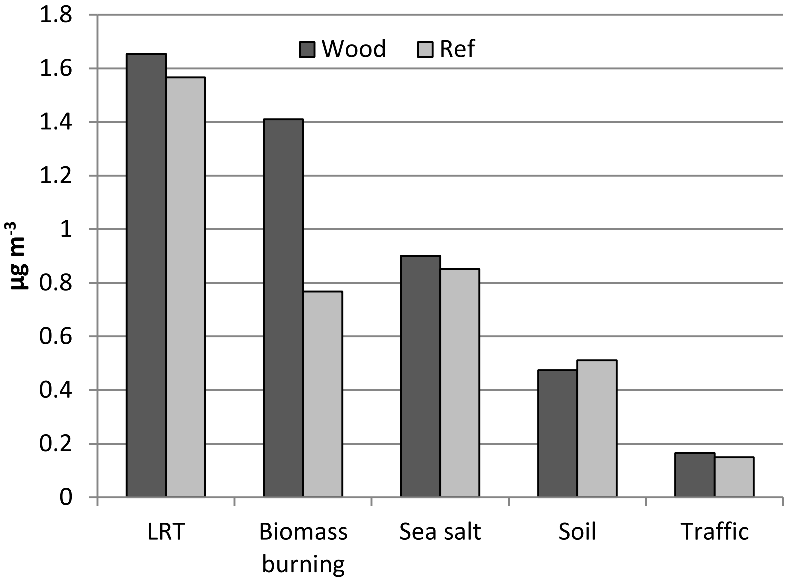

Figure 3. Source contribution in the wood-burning area and at the reference station from the five-factor PMF model.