



**Characterization of particles in
Lycksele and Gothenburg**

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1. Abstract

In some small towns in Sweden, the concentration of particles can sometimes increase to extreme levels, especially during stable meteorological conditions. The towns are characterised by a large extent of residential wood burning, beside other local particle sources such as vehicle exhaust, road traffic resuspension and long-distance transport.

Lycksele in Northern Sweden was chosen to represent a typical small town characterised by residential wood burning. Gothenburg in Southern Sweden represented a site dominated by road traffic. During a winter-period, the amount of particulate PAH per unit particulate mass $<2.5 \mu\text{m}$ in diameter (PM_{2.5}), was larger in Lycksele than in Gothenburg. It was also found that five PAHs, known for their carcinogenicity, were present in higher concentrations in Lycksele than in Gothenburg during the winter-period. During spring the concentration in Lycksele was lower than in Gothenburg. The concentration in Gothenburg did not change between the periods.

The sources to PM_{2.5} in Lycksele was estimated using two independent source-receptor methods, principal components analysis (PCA) and positive matrix factorization (PMF). The average contribution to PM_{2.5} from wood burning during the winter-period was estimated to range between 9-34%. The particle contribution from wood burning during extreme cold winter nights could be as high as 70%. The average contribution to PM_{2.5} from wood burning was similar to the contribution from road traffic, using the PMF-method, but 25-33% of the road traffic contribution using the PCA-method.

During the spring period, the average contribution from resuspension to PM_{2.5} in Lycksele was estimated to be about 70%.

The PAH-data from the winterperiod in Lycksele was analysed using three multivariate methods, PCA, PMF and chemical mass balance (CMB). The contribution from road traffic to total mass PAH was similar to the contribution from wood burning.

2. Introduction

High levels of particles in the ambient air are believed to be health hazardous and cause respiratoric problems. It is not known whether it is the adsorbed chemical compounds or the physical characters of the particles that are the important variables, or a combination of the two.

In small Swedish cities the amount of particles can increase to high concentrations in wintertime due to emission from local sources combined with stable meteorological conditions. The concentration can then be as high as measured in large cities in Southern Sweden (Areskoug et al., 2001). Likely local sources to the particle concentration are traffic exhaust, resuspension and residential wood burning.

This study is a part of a national effort to measure the concentrations of PM and different sources to PM in Swedish cities. The PM₁₀ and PM_{2.5} concentrations observed in 5 cities and at 2 background sites have been presented elsewhere (Areskoug et al. 2001). This study focuses on the estimates of the contribution to PM and PAH from different sources. The most important source contributions to the particle concentration is estimated by using different multivariate techniques. For this study, a town in northern Sweden, Lycksele, was chosen to represent a community where a large fraction uses wood burning for residential heating, and where the meteorological conditions can cause the particle concentration to increase to high, sometimes extreme, levels. The sampling site was situated in the outskirts of the city core, near a river that passes through the town. No tall buildings were placed nearby. The site was situated about 50 meters from E12, an European highway, where 11 000 vehicles per day passes.

Samples were also collected at a street level site (Gårda) in Gothenburg during the same period. The samplers were placed at an open site about 5-10 meters from E6, a four-laned busy road, where 79 000 vehicles passes per day. For some comparisons, samples collected at a background site, Aspvreten, have been used. Aspvreten is situated about 100 km south of Stockholm.

3. Experimental

During two field campaigns in spring 2000, 40 samples from each site were collected and analysed regarding volatile organic carbons (VOC), hydrocarbons (alkanes) and polycyclic aromatic hydrocarbons (PAH, see Table A1). In addition, samples for metal and elemental analysis were collected during January-April 2000. About 50 samples from Lycksele, from fine and coarse mode, have been analysed, as well as 17 fine mode samples from Gothenburg. The samples were collected during 12 hours. The PAHs were analysed by Miljölaboratoriet in Nyköping, using gas chromatography-mass spectrometry (GC-MS). The analysis of elements was performed at the department of Nuclear Physics, Lund University, using Proton Induced X-ray Emission (PIXE). The VOCs were analysed on GC-MS at ITM, Stockholm University.

The first period of the field campaign took place between January 25 - February 4 and is referred to as “January” or period 1 samples. The second period started April 3 and ended April 14. It is referred to as “April” or period 2 samples. The periods for the elements are somewhat different, but referred to as “January” and “April” anyway.

During the period of sampling, continuous measurements of PM_{2.5} and PM₁₀ were made. However, for the campaign periods only PM_{2.5} was measured in Lycksele.

4. Concentrations of pollutants

4.1 Total concentrations

4.1.1 Lycksele

During the campaign in January-February in Lycksele it was seen that the amount of total PAH increased as PM_{2.5} increased (Figure 1a), and this occurred during periods when the temperature decreased to levels below -10°C. The temperature dependence was not as obvious in the campaign during April (Figure 1b). Yet it can be seen for two samples when the temperature decreased during night increased with respect to the sum PAH. It is likely that the periods of decreased temperature leads to increased emissions of pollutants due to

residential heating, in this case increased emissions of PAH (Figure 1c). In January, the correlation coefficient between temperature and concentration of sum PAH was $R^2=0.67$, while there was no correlation between temperature and sum PAH in April ($R^2=0.05$). The lack of correlation in April may be due to the fact that the temperature range was not very large and there was only one short period with temperatures below -10°C . The increase of PAH levels in January may also have been enhanced by stable meteorological conditions that prevented dilution of the pollutants.

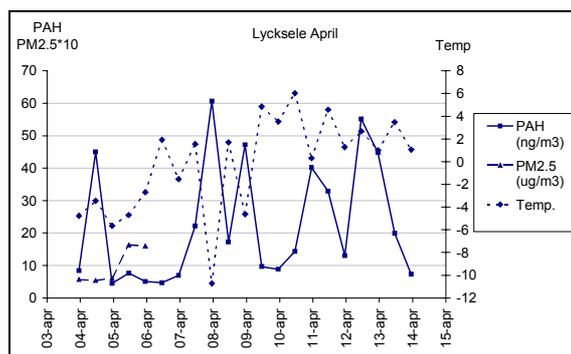
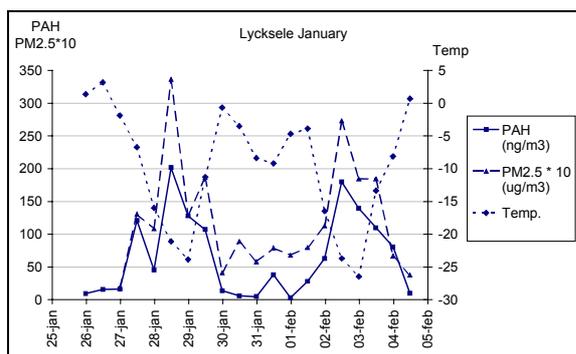


Figure 1a. Concentrations of PAH and PM2.5 during the campaign in January in Lycksele. The temperature is also presented in the plot.

Figure 1b. Concentrations of PAH and PM2.5 during the campaign in April in Lycksele. The temperature is also presented in the plot. Only a limited amount of PM2.5-data was sampled.

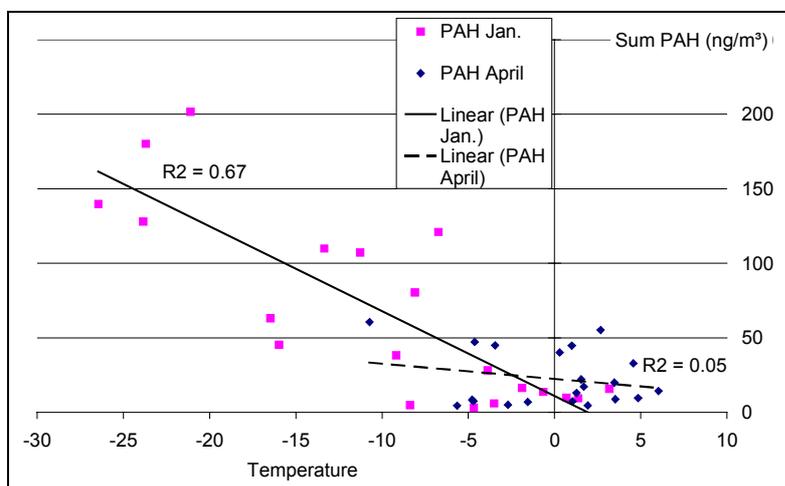


Figure 1c. PAH-concentrations plotted to the temperature in Lycksele in January and in April. The R^2 -values are also presented.

The correlation coefficient between the concentrations of PAH and PM2.5 in January was $R^2=0.83$. During the campaign in April there was too few PM2.5-data to be able to compare with PAH.

Comparing the emission of VOC¹ and PAH (Figure 2a and b) showed that these groups of chemicals varied in the same way both in January and in April ($R^2=0.72$ and $R^2=0.74$, respectively), which indicates that the sources responsible for the emissions could be the same.

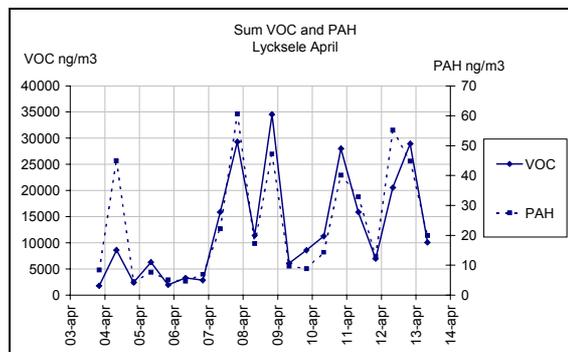
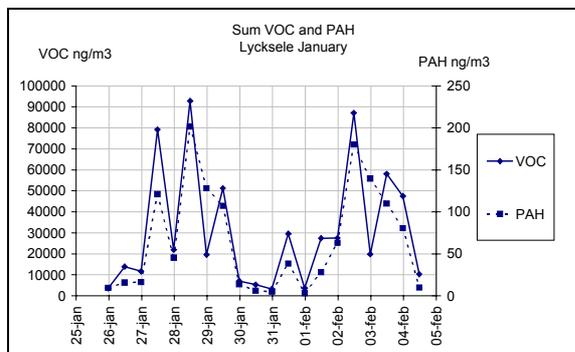


Figure 2a. Measured concentrations of sum VOC and PAH in Lycksele in January ($R^2=0.72$).

Figure 2b. Measured concentrations of sum VOC and PAH in Lycksele in April ($R^2=0.74$).

4.1.2 Gothenburg

There was a weak correlation between the PAH and PM_{2.5} in Gothenburg ($R^2=0.41$, Figure 3a), but the concentrations showed no temperature dependence, which was seen in Lycksele. The character of the major source to PM_{2.5} at Gårda in Gothenburg, resuspension, is not expected to be temperature dependent. However, it should be noted that the temperature range during the period was very small (+5 to -1°C).

In figure 3c it can be seen that the concentrations of VOC correlate with PAH in Gothenburg in April ($R^2=0.64$). There were not enough VOC-data during January to make a comparison.

¹ The sum of VOC is from benzene – 2-mehtylnaphtalene from table A1

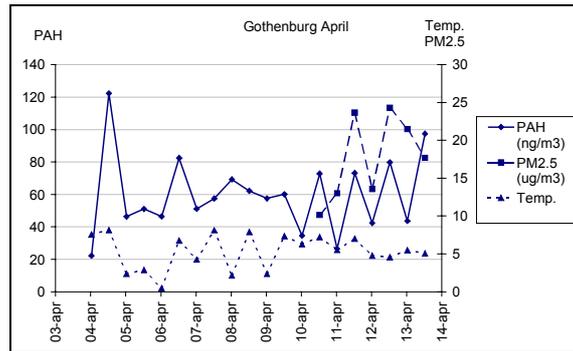
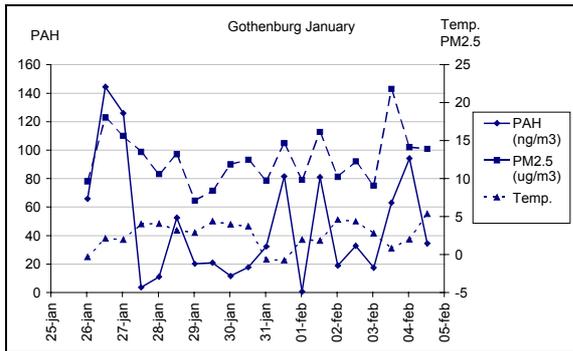


Figure 3a. Concentrations of PAH and PM2.5 during the campaign in January in Gothenburg. Temperature is also presented in the plot.

Figure 3b. Concentrations of PAH and PM2.5 during the campaign in April in Gothenburg. Temperature is also presented in the plot. Only a limited amount of PM2.5-data was sampled.

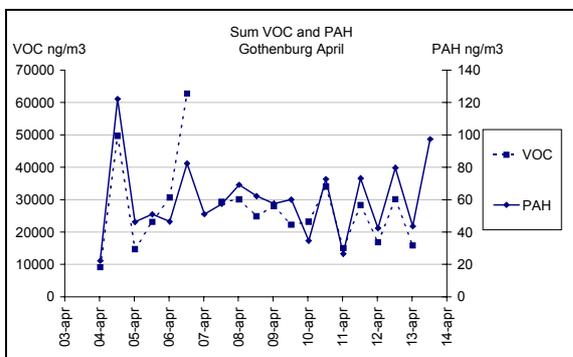


Figure 3c. Measured concentrations of VOC and PAH in Gothenburg in April.

4.2 Comparison of chemical composition

A major part of the pollutants in Gårda in Gothenburg is expected to derive from road traffic. The pollutants in Lycksele are expected to be a mix of pollutants from residential wood burning and road traffic. The chemical composition of particles in Gothenburg is therefore expected to be different from Lycksele. The concentrations of PAHs were examined to evaluate this.

Many PAHs are semi-volatile, which means that they may be present in the atmosphere both as gases and adsorbed to particles. Low vapour pressure of a compound and high particle concentration in the air favour adsorption to particles. In general, the vapour pressure decreases with increasing molecular weight. Further, the vapour pressure increases with

increasing temperature. In Table A1, the PAHs are arranged due to volatility. Acenaphthylene is mostly gaseous and coronene is exclusively found on particles.

In this study, gaseous and particle-bound PAHs were sampled together. Therefore, their concentrations on particles could only be estimated. To do this, the particle concentration must be known and we had to assume that the system was in equilibrium. By the design of the sampler and the airflow, we assumed that the size of the particles sampled was equivalent to PM2.5, which was measured during the campaign.

Figure 4 shows the amount particulate PAH/ $\mu\text{g PM}_{2.5}$ for Lycksele and Gothenburg for both periods. It was assumed that all PAH from benzo(a)fluorene and heavier (see Table A1) was particle bound. The line represents the measured background value from Aspvreten. It can be seen that in Lycksele in January the amount PAH/PM2.5 was about double the amount in Gothenburg, but in April there was no difference. For both places and periods the amount was above the background value. The local sources lead to a 4 – 10 times increase of the particulate amount of PAHs.

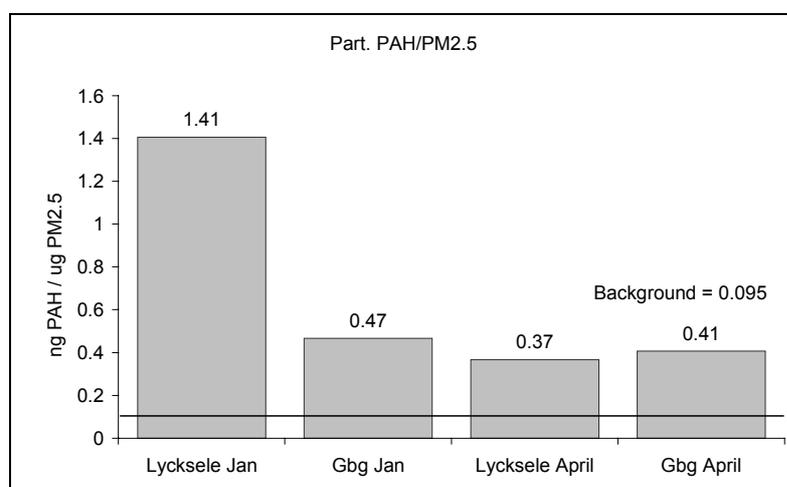


Figure 4. Sum of particulate PAHs (benzo(a)pyrene – coronene) in ng per $\mu\text{g PM}_{2.5}$. It can be seen that there are more PAHs on the particles in Lycksele in January than in April and in Gothenburg.

Another way of estimating the particle concentration of PAHs, is to use the relations between a) the subcooled liquid-vapour pressure of each compound and temperature and b) this vapour pressure and the fraction being particle bound. For 13 of the 23 measured PAHs, corresponding to 70 % of the total PAH mass sampled, the vapour pressures were calculated

according to Yamasaki et al (1984), cited in Pankow and Bidleman (1992). The average temperature for each 12 h sampling occasion was used. The fraction particle bound was then calculated according to Equation 5 in Finizio et al. (1997).

In Gothenburg the temperature and PM_{2.5} were relatively constant during the January campaign and so was the fraction particle-bound PAH. Only about 20 % of phenanthrene, a fairly volatile compound, was particle-bound, while 80 % of the less volatile benzo(a)fluorene was adsorbed to particles. Still, phenanthrene was one of the most abundant compounds bound to particles, due to its high total air concentration.

Temperature and PM_{2.5} varied strongly in Lycksele in January. The concentration of PM_{2.5} was highest during the periods with the lowest temperatures and thus both parameters contributed to high fractions of particle-bound PAHs. When the temperature decreased below -20°C , the particle-bound fraction was 100 % for the least volatile PAHs and about 40 % also for the more volatile PAHs. Phenanthrene was the most abundant PAH on particles during the cold periods.

This way of estimating the fraction particle-bound PAH, gave a higher particle concentration of PAH, calculated as ng PAH/ μg PM_{2.5} compared to what was showed in Figure 4, but the relation between the two sampling stations/measuring campaigns was similar.

Of the particulate PAHs measured, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene and dibenzo(a,h)anthracene are of special interest due to their carcinogenic properties. In Figure 5a, the concentrations of these compounds are compared between the two sites and periods. The amounts measured in Lycksele in January were higher (both average and median values) compared to April. The concentrations in Gothenburg did not differ between the periods. The average concentration in Lycksele compared to Gothenburg in period 1 was 3-5 times higher. For the second period, the concentrations measured in Gothenburg were 2-3 times higher than in Lycksele.

In Figure 5b, the fraction of the carcinogenic compounds per total mass PAH is shown. It can be seen that the relative amount of the carcinogenic compounds is larger in Lycksele during the winter-period than during spring. The relative amount in Lycksele during the first period is also larger than the relative amounts measured in Gothenburg.

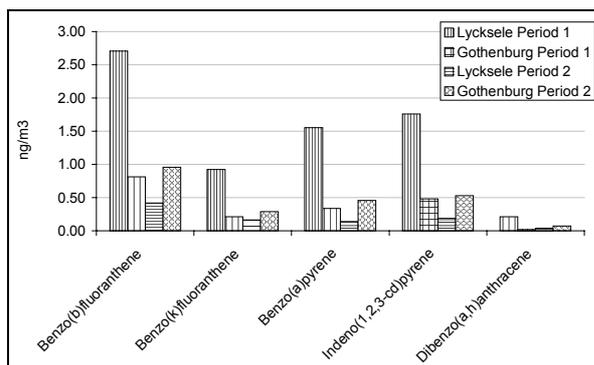


Figure 5a. Average concentrations of carcinogenic, particulate PAHs in Lycksele and Gothenburg during the two campaign periods. It can be seen that the concentrations were highest in Lycksele in January and lowest in Lycksele in April. The concentrations in Gothenburg did not differ between the periods.

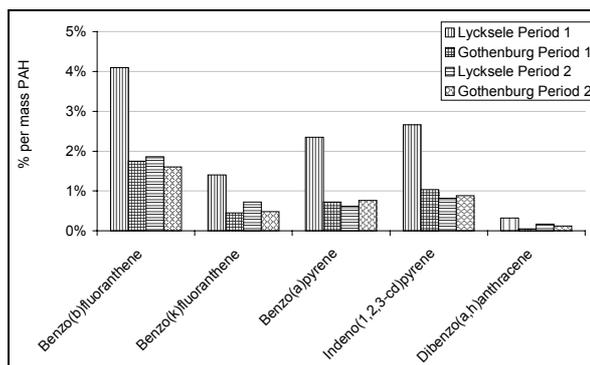


Figure 5b. Average fractions of carcinogenic, particulate PAHs in Lycksele and Gothenburg during the two campaign periods, in % of compound of total mass PAH. The carcinogenic part of PAH is larger in Lycksele in January than in Gothenburg. In April in Lycksele, the fraction is about the same as in Gothenburg.

In table 1, the concentrations of different groups of compounds and PM2.5 are presented for both places and periods. It is difficult to compare absolute concentrations of different compounds observed in Gothenburg and Lycksele due to the short time period of sampling. However, it may be noted that the ratio of alkanes (predominantly from vehicle exhaust) to PAH (in Lycksele a mix of vehicle exhaust and wood burning) is much higher in Gothenburg as compared to Lycksele. During period 1 the ratio is 18 and 78 ng alkanes/ng PAH in Lycksele and Gothenburg, respectively. During period 2 the ratios are more similar, 40 and 47 respectively. This indicates that there are different sources in Lycksele compared to Gothenburg. The difference in relative concentration of different compounds between Lycksele and Gothenburg could also be due to different background concentrations of these compounds.

Table 1. Summed contribution from different chemical groups (compounds included are presented in Table A1, except for alkanes that includes C10-C20). Comparisons between Gothenburg and Lycksele for periods 1 and 2.

MAX	1		2	
	Lycksele	Gothenburg	Lycksele	Gothenburg
ng/m ³				
VOCs	95 000	27 000 ¹	37 000	63 000
Alkanes	2 800	2 800 ¹	2 300	10 000
PAHs	200	144	60	122
PM2.5 (µg/m ³)	34	22	16 ²	24 ³

MEDIAN	1		2	
	Lycksele	Gothenburg	Lycksele	Gothenburg
ng/m ³				
VOCs	23 000	23 000 ¹	10 000	26 000
Alkanes	750	2 500 ¹	550	2 700
PAHs	40	32	14	58
PM2.5 (µg/m ³)	10	12	6 ²	18 ³

¹ Based on four samples

² Particle measurements for 1/4 of the period only

³ Particle measurements for 1/3 of the period only

5. Source characterisation

5.1 Estimations of source contributions - Methods

There are many ways of estimating the contribution from different sources to a sampling site. Different multivariate techniques have been used in this work. The results have been compared with results derived from simpler source estimations.

5.1.1 Chemical Mass Balance – CMB

The CMB is a least-squares solution to a set of linear equations that explains the receptor concentration as a sum of source contributions and source compositions (Watson et al., 1991). The sources contributing to the receptor site has to be known and quantified to be able to use this method.

The method was more useful when applied on PAHs than PIXE-elements. Since PIXE-elements could be quantified using other techniques (PCA and PMF), no further effort was put into solving the CMB quantification problem for that data set.

One disadvantage using the CMB-method is that if one or more sources are unknown, the model cannot detect this, and will model the mass from the unknown source to a known.

5.1.2 Principal Components Analysis – PCA

The PCA method is applied on the data matrix of the receptor site (samples \times variables (compounds)) to reduce the matrix so that a minimum number of factors (components) can explain the maximum variance in the data set (in terms of scores and loadings) (explained in e.g. Bro (1996)). The scores explain the temporal variation of the factors, and the loadings explain the variables correlation with the factors (Figure 6). The method requires many more samples than variables in order to get a good source estimation and a statistically significant result. The derived factors are orthogonal and can be orthogonally rotated for a better interpretation. The most widely used rotational technique is the so-called Varimax-rotation, which was applied to the data set used here. One disadvantage of PCA is that the components derived may be negative, and in this case, there is no physically meaningful interpretation of negative source contribution.

Absolute PCA (A-PCA) is the transformation of scores and loadings to absolute measures, i.e. concentrations due to each source (Swietlicki et al., 1996). The PCA method requires prior knowledge about relative source compositions in order to interpret the components correctly. The scores derived may be used to calculate the temporal variation of e.g. PM_{2.5} from different sources by using least-squares approximation (multiple linear regression – MLR).

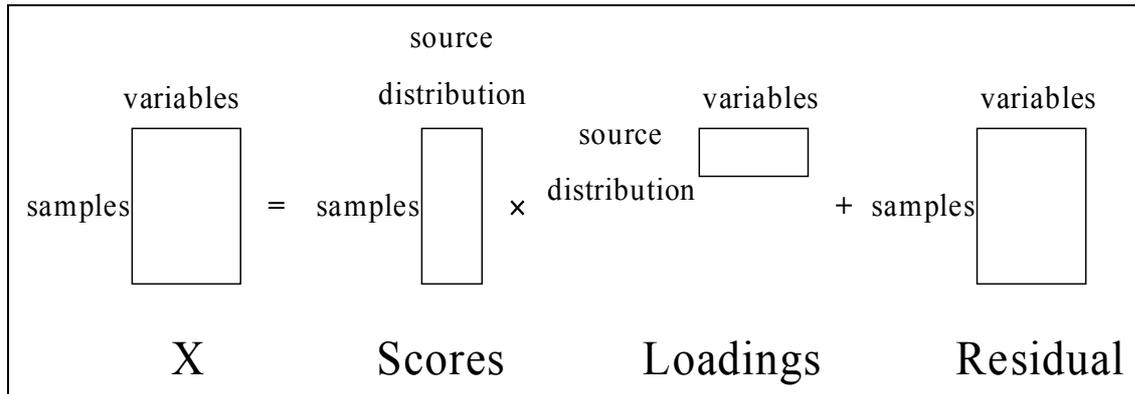


Figure 6. The partitioning of the data matrix (X) into scores and loadings. The residual consists of the unexplained part of the data matrix.

5.1.3 Positive Matrix Factorisation – PMF

PMF is a new multivariate technique developed by P. Paatero of the department of Physics, University of Helsinki, Finland (Paatero and Tapper, 1994). One of the largest benefits of using PMF compared to PCA is the non-negativity constraint that is included in PMF. Other benefits are that the components (sources) do not have to be orthogonal (independent), and when modeling, a matrix of standard deviations is included so that samples, variables or values with a high uncertainty can be weighted in the model, i.e. more reliable samples will have a larger influence than less reliable samples. Missing values can also be treated, thereby no samples or variables has to be excluded in order to fit a model.

In the same way as for the PCA-method, PMF can be used with MLR to calculate the temporal trends in PM2.5 concentrations due to different sources.

5.1.4 “Sum of oxides” – method

By using the sum of oxides method, the assumption is made that road dust is mainly composed by the oxides of six elements: SiO₂, Al₂O₃, Fe₂O₃, TiO₂, CaO and K₂O. The mass concentration of dust is given by the equation:

$$\text{Road dust} = 2.14 \cdot \text{Si} + 1.98 \cdot \text{Al} + 1.43 \cdot \text{Fe} + 1.67 \cdot \text{Ti} + 1.4 \cdot \text{Ca} + 1.2 \cdot \text{K}$$

where Si, Al, Fe, Ti, Ca and K are the measured concentrations (Andrews et al., 2000).

5.2 Estimations of source contributions – Results from Lycksele

5.2.1 Elements

Metals and other elements are commonly used for source-receptor studies of particles. Samples are collected on filters. The composition of the compounds in different source matrices is fairly well known. Most elements do not undergo reactions during transport in air and thereby the relative composition of the compounds will not change compared to the source composition.

Five significant sources were found using PCA on all data. To increase the degrees of freedom, compounds known to the sources were kept and other compounds excluded. Still five significant sources were found (Table A2). Three of the loading values in the first four components were negative by a small amount, and were therefore replaced with zero. In the fifth loading, all elements except S and Br had negative values and were set to zero.

For PMF, four components gave the best model (Table A3). The same PIXE elements were used for both PCA and PMF.

By using the components variation in time compared to the variation of PM_{2.5} in time, a MLR could be used to estimate the source contributions to PM_{2.5}. PCA and PMF gave somewhat different results (Table 2). The largest difference between the results from PCA and PMF was the contribution from resuspension. It was smaller during January using PMF compared to A-PCA, 25 and 55% respectively. In April the methods modeled the contribution to be the same, about 70%. Also the wood burning component increased during cold periods in January using PMF. This is probably due to the fact that the components in PMF not necessarily have to be orthogonal as they have to in PCA.

The average contribution from wood burning to PM_{2.5} using PMF was estimated to be 18%, and the resuspension contribution was 53%. The contribution from traffic was similar to that of wood burning, and there was a long-distance component that contributed about 11% to

PM2.5 in average. During cold periods with inversion, the contribution from wood burning to PM 2.5 was found to be as large as 70% (Figure A1).

Table 2. Source contributions to PM2.5 (%) using different techniques on PIXE elements in Lycksele.

	Wood Burning	Road Traffic (directly)	Resuspension	Uncertain (possibly local)	Long-distance contr.	Explained Mass (Calc/Meas) ¹
A-PCA	6	24	63	1	5	94
<i>January</i>	9	30	55	1	6	81
<i>April</i>	5	21	70	1	4	106
PMF	18	18	53	-	11	86
<i>January</i>	34	34	25	-	7	74
<i>April</i>	7	7	72	-	13	97
(NH ₄) ₂ SO ₄					2-11	
Sum of Oxides-method			4-10			

¹ Calculated mass / Measured mass

5.2.1.1 Detailed description of the components

One large component seems to be road dust, with high loadings on e.g. Si and Ti (Table A2 and A3). Compounds known to be relatively abundant in wood smoke (K and Ca) had large loading values in this component too, which indicates that it might be a mixed source. What speaks for the earth crust-contribution is that the compounds included in the component are typical earth crust compounds as Si and Ti. In Lycksele, the contribution from the resuspension component ranged from 53-63% depending on which method was used. If the source contributions were divided into the two different periods (January and April) for the PCA method, the contribution from resuspension was 55 and 70% respectively. Even in the fine particle size fraction (<2.5 μm), as much as 70% can come from resuspension when there is low humidity in air, as shown by Isaksson (2001) for Stockholm. As for the period in

January, the ground in Lycksele was covered by snow and the roads by ice, and a contribution of 55% from resuspension seems unreasonably large. When dividing the source contribution from the PMF method into the two periods, the contribution from resuspension was 25% and 72% of the PM_{2.5} concentration in January and April respectively. The fraction of resuspension seems more reasonable for period 1.

A large contribution from resuspension was modeled for March 25-27 during nights using PMF (Figure A2). A similar contribution from resuspension was also found using PCA. Areskoug et al. (2001) showed that the contribution to particulate mass during this period was extreme at 19:00-20:00, four evenings in a row (March 25-28). These high PM_{2.5} levels occurred when the temperature decreased. The regularity in the extreme particulate levels and the time when it occurred suggest that the source might be wood burning rather than resuspension. This seems consistent with the fact that the elements in the component (Table A2 and A3) suggest it to be a mixed source of both wood burning and resuspension.

Using the sum of oxides method on the fine fraction in Lycksele, an average dust contribution to PM_{2.5} was found to be about 4%. The maximum value obtained using the oxide method was 10%. This much lower than the 25-72% modeled by the PCA and PMF approach. So far, we have no explanation for these different results.

The second largest component seems to be a traffic-related component, with high loadings on Cu and Pb. The PCA modeled this component to contribute about 24% to PM_{2.5} and PMF modeled it to 18%.

The third large component seems to be a wood burning component. K and Zn are the compounds known to be tracers for wood burning aerosols. The pattern seen in Hedberg et al. for birch wood (Table A4) agrees well with the pattern found in both PCA and PMF absolute loadings (Table A2 and A3). This component contribute 6-18% in average to PM_{2.5} mass. PMF models the contribution from this component to be as large as the contribution from road traffic.

The fourth component is a long-distance component, as can be seen because of the S and Cl. The contribution in time agrees with the expected for a long-distance component, i.e. it is relatively constant. The sources might be sea-salt particles (consists of e.g. Cl) mixed with

pollution from e.g. the Kola Peninsula, which is a large emitter of S. The contribution to PM_{2.5} was 5% using PCA and 11% using PMF.

The fifth component for PCA has high loadings on S and Ni, which indicates that it corresponds to oil combustion. A sequential time plot shows that this component was nearly the opposite of the temperature, indicating that the component derives from local combustion. Another interpretation might be long distance transport, as sulfur typically is higher during such periods. The contribution to PM_{2.5} from this component was 1%. This component was not found when modeling using the PMF method.

If the assumption is made that all S measured in Lycksele is due to long-range transport in the form of (NH₄)₂SO₄, an approximation of the contribution from long-distance transport to PM_{2.5} can be made. The average contribution was then 2% and the maximum value obtained was 11%. This range lies within the models prediction.

5.2.2 PAH

A Varimax-rotated PCA was applied on the concentrations of PAHs in Lycksele in January, and two components were extracted from the data set. By studying the components relative importance in time, and assuming that retene is a tracer mainly of softwood burning, the identification of the likely sources could be made. However, the contribution from the two sources, wood burning and traffic, only explained 57% of the measured mass (Table 3). The unexplained part might be the average background contribution. This would be the same contribution from the background as obtained using the CMB-method.

The PAH data for January was also analyzed using PMF. The sources derived from the analysis were wood burning and traffic, and explained 43 respectively 57 % of the explained mass. The relative contribution from wood burning to total mass PAH is the same using all the three methods, i.e. about 40%.

The data for January was easier to interpret than the data for April, using PAHs. PAHs known to be emitted in traffic exhaust were separated from wood burning tracer, retene, among others during the first period, while during the second period, the components do not

show any source specific pattern at all. If the number of samples collected had been larger, an interpretation for the spring period probably would have been possible.

Table 3. Source contributions (%) to summed mass of PAHs using different techniques on PAHs in Lycksele.

	Wood Burning	Road Traffic	Long-distance transport	Explained mass (Calc/Meas)
CMB	35	25	40	
A-PCA	40	60	-	57
PMF	43	57		96

5.3 Simple source characterization in Gothenburg using PIXE-elements

Using the sum of oxides method, mentioned above, to estimate the relative contribution to particle mass from dust, the results varied from 2-20% contribution. The average value was 7.5%, which was twice the fraction measured in Lycksele. Since there were only limited amount of data from Gothenburg, the fraction dust may not be representative of the actual distribution, but gives a hint that it is larger than the measured amounts in Lycksele.

In the same way as was done for Lycksele, the contribution of long-range transported particles using S was calculated for Gothenburg. In Gothenburg the average contribution was 5.8%, and the maximum amount measured was 11.4%. This was less than expected, but also here it must be emphasized that the number of samples was low, and that the results might not be representative for the contribution over a longer period.

6. Concluding discussion

The amount particulate PAH per PM_{2.5}, was larger in Lycksele than in Gothenburg. A larger fraction of PAH on particles in Lycksele is consistent with an additional local source of PAH there. A larger fraction of other particulate compounds than PAH in Gothenburg would give the same result, but would also point to differences in sources at the two sites.

It was also seen that five PAHs, known for their carcinogenicity, were present in higher concentrations in Lycksele than in Gothenburg during the winter period. During spring the concentration in Lycksele was lower than in Gothenburg. The concentration in Gothenburg did not change between the periods. The abundance of these carcinogenic compounds was also higher during the winter period in Lycksele, with respect to total concentration of PAHs, than in spring and in Gothenburg.

The sources to PM_{2.5} in Lycksele was estimated using two independent source-receptor methods, principal components analysis (PCA) and positive matrix factorisation (PMF). The average contribution to PM_{2.5} from wood burning during the winter-period was estimated to range between 9-34%. The contribution from wood burning during extreme cold winter nights could be as high as 70%. The average contribution to PM_{2.5} from wood burning was in the same range as the contribution from road traffic, using the PMF-method, but 25-33% of the road traffic using the PCA-method.

During the spring period, the average contribution from resuspension was estimated to be about 70% of the PM_{2.5} concentration in Lycksele.

The PAH-data from the winter-period was analysed using three multivariate methods, PCA, PMF and CMB. All three methods showed that the average contribution from wood burning to the total concentration of PAH was similar to the contribution from road traffic.

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9. Appendix

Table A1. Chemical parameters sampled and analyzed in Lycksele and Gothenburg during the campaigns in spring 2000.

PAH	VOC	Alkanes	Elements (PIXE)	Other parameters
Acenaphthylene	Benzene	Nonane	Al	PM2.5
Acenaphthene	Toluene	Decane	Si	PM10
Fluorene	Ethylbenzene	Undecane	P	Meteorological parameters
Phenanthrene	m-xylene	Dodecane	S	
Anthracene	o-xylene	Tridecane	Cl	
2,6/3,5-dimethylphenanthrene	1,3,5-trimethylbenzene	Tetradecane	K	
1,7-dimethylphenanthrene	1,2,4-trimethylbenzene	Pentadecane	Ca	
Fluoranthene	1,2,3-trimethylbenzene	Hexadecane	Ti	
Pyrene	Naphtalene	Heptadecane	V	
Retene	1-methylnaphtalene	Octadecane	Cr	
Benzo(a)fluorene	2-methylnaphtalene	Nonadecane	Mn	
Benzo(b)fluorene	m-ethyltoluene	Eicosane	Fe	
Benzo(a)anthracene	p-ethyltoluene		Co	
Chrysene	o-ethyltoluene		Ni	
Benzo(b)fluoranthene			Cu	
Benzo(k)fluoranthene			Zn	
Benzo(e)pyrene			Ga	
Benzo(a)pyrene			Ge	
Perylene			As	
Indeno(1,2,3-cd)pyrene			Se	
Benzo(g,h,i)perylene			Br	
Dibenzo(a,h)anthracene			Rb	
Coronene			Sr	
			Zr	
			Mo	
			Pb	
			Y	
			Nb	

Table A2. Absolute loadings obtained using PCA (ng/m³).

	Resuspension	Road Traffic	Wood burning	Long-distance transport.	Uncertain (ev. local oil comb.)
Si	136.8	66.3	30.8	0.0	62.6
S	4.1	10.1	9.1	12.4	62.0
K	23.0	11.5	49.3	0.0	15.0
Ca	13.2	5.3	2.4	0.0	4.2
Ti	3.4	0.9	0.0	0.0	1.3
Mn	0.6	0.4	0.6	0.0	0.3
Fe	36.9	26.8	7.0	0.0	14.0
Co	0.1	0.3	0.0	0.0	0.1
Ni	0.0	0.0	0.0	0.0	0.1
Cu	0.2	1.0	0.6	0.0	0.0
Zn	0.3	2.0	6.9	0.0	1.2
As	0.1	0.1	0.0	0.0	0.1
Br	0.0	0.0	0.0	0.7	0.0
Pb	0.0	0.5	0.2	0.0	0.0
Contr. to PM2.5 (%)	63	24	6	5	1

Table A3. Absolute loadings obtained using PMF (ng/m³).

	Resuspension	Road Traffic	Wood burning	Long-distance transport.
Si	251.0	0.0	6.9	0.0
S	4.5	1.6	14.2	23.6
K	35.5	0.0	24.7	2.1
Ca	23.5	0.9	0.9	1.9
Ti	5.5	0.0	0.0	0.1
Mn	1.0	0.2	0.2	0.2
Fe	66.1	0.4	1.5	0.6
Co	0.0	0.1	0.0	0.1
Ni	0.0	0.0	0.0	0.1
Cu	0.3	0.6	0.1	0.0
Zn	0.1	0.5	3.6	0.0
As	0.2	0.0	0.0	0.0
Br	0.0	0.1	0.0	0.3
Pb	0.0	0.2	0.0	0.0
Contr. to PM2.5 (%)	53	18	18	11

Table A4. Source composition of particles emitted from burning birch wood in a wood stove during an emission study (Hedberg et al).

Compound	Relative Abundance (%)
Si	15.6
S	11.0
Cl	9.6
K	46.2
Ca	0.7
Cr	1.1
Mn	0.2
Fe	0.4
Co	0.1
Ni	0.1
Cu	0.1
Zn	12.7
Se	0.0
Br	0.3
Rb	0.3
Mo	1.1
Pb	0.6

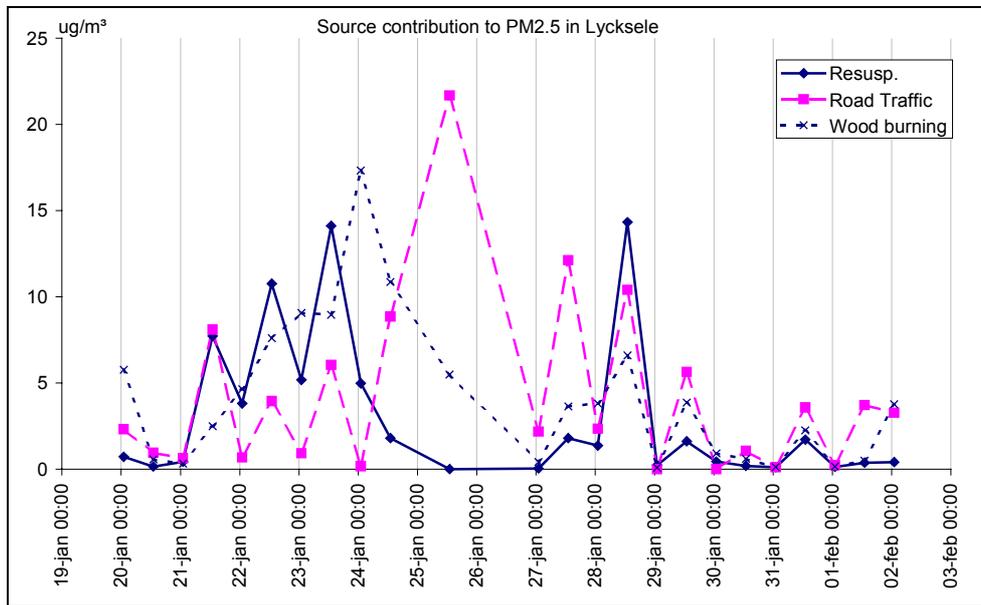


Figure A1. The estimated contribution from resuspension, road traffic and wood burning to PM2.5 in $\mu\text{g}/\text{m}^3$ in January in Lycksele using PMF.

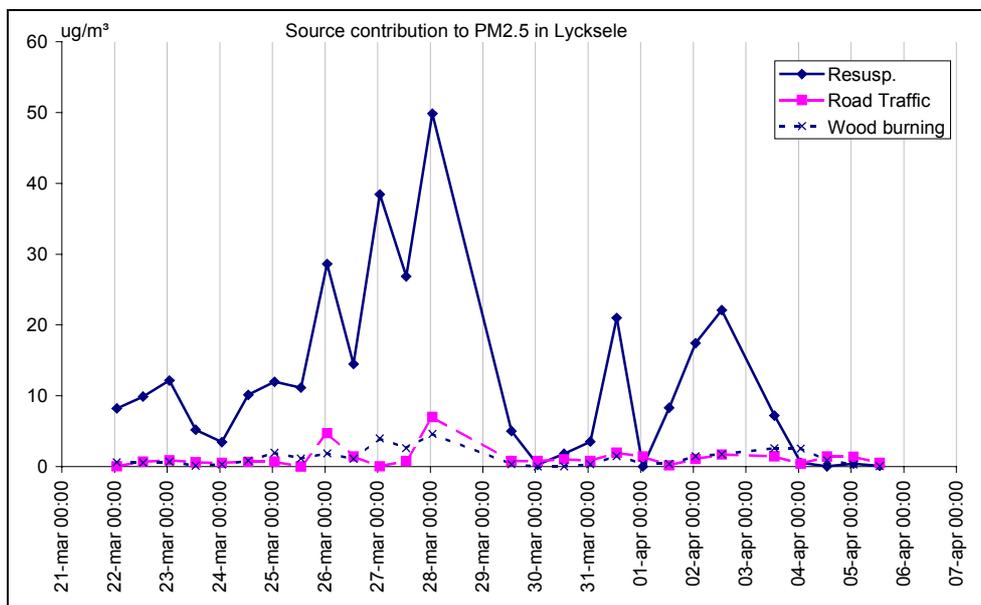


Figure A2. The estimated contribution from resuspension, road traffic and wood burning to PM2.5 in $\mu\text{g}/\text{m}^3$ in April in Lycksele using PMF.



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