Discriminant Analysis of Heavy Metal Concentrations in the Soil of St. John's, Newfoundland

Stacy Campbell

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Dr. Schneider
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Introduction

Metals are widely distributed in the environment, occurring naturally in bedrock, soil, water and air. However recent anthropogenic additions to the environment have caused concentrations to rise to a level of concern. Mining wastes, industrial emissions, land application of sewage sludge, fossil fuel combustion, and the production of batteries, alloys, and other industrial products all contribute to the concentration of metals in our environment.

The contamination of soil with heavy metals is a health concern, with many negative health effects associated with compounds such as arsenic and lead, the latter being of particular concern in St. John's. Lead is cumulative poison that has well known negative health impacts on humans, especially on children. Chronic low-level lead poisoning can cause neurological and developmental problems leading to decreased cognitive abilities. Research conducted in the last 30 years has indicated that soil lead concentrations are generally higher in urban environments that display an older housing stock containing leaded paint, a high concentration of industry, and heavy traffic (Mielke 1994). Since lead does not biodegrade and is highly immobile in soils, concern about soil contamination persists despite the fact that most lead was removed from residential paints in the late 1970's and from gasoline in 1990.

The preliminary investigation into the level of lead in urban soil in St. John's was prompted by results from an earlier study on lake sediment chemistry in the local area (Christopher 1999). Sediment cores were found to be rich in many heavy metals, with lead levels increasing thirty times in the last 200 years. It was suggested that the high levels of lead were due first from the combustion of coal, and then later from leaded gasoline emissions. The author also hypothesized that since these sources of lead cause widespread pollution that the sediment core findings might not be limited to aquatic environments, but that high metal concentrations might also be present in surface soil.

In response a pilot soil sampling program was initiated in the summer of 2003 with the broad objective of determining whether soil in St. John's contains high levels of heavy metals, particularly lead, and to see if the sources of contamination could be discovered through a planned sampling program targeting specific land uses.

This paper will address one of the project's sub-objectives, determination of the sources of lead in the contaminated soil. Of the potential sources, the deterioration of exterior leaded paint, the combustion of leaded gasoline, and the burning of coal in home heating systems were determined to be of most concern in St. John's. High lead is therefore predicted along the foundations of older homes, beside roadways, and in the backyards of houses where coal ash may have been dumped. If these sources can be confirmed, then it will be easier to spatially predict areas of concern within the city and on individual properties.

Each of the sources of lead may contain additional metals, some of which may be unique to that type of contamination. A quick review of the literature has found that paint often has high levels of zinc, cadmium, manganese, nickel, copper, cobalt, chromium, and vanadium (Mielke et al 2001), while copper, zinc, molybdenum, and barium have been linked to roadside pollution (Harrison et al 2003). The metal content of coal is of specific to the area where it was mined, but often includes arsenic, zinc, mercury, and antimony, along with others (Block and Dams 1976). If we analyze the metal concentrations of samples predicted to be mainly influenced by a single source of pollution using discriminant analysis we may be able to find the signature metal composition for each source. Then any future samples could be entered into the discriminant analysis equation and classified as to dominant pollution source.

Methods

Sample Collection and Analysis

Soil samples were collected from four different categories of land use: open space, parks, school property, and residential property. Sample sites were selected to represent a broad range of land uses in the city, although the majority were collected downtown as that was predicted to be the area of most concern. Soil samples were also collected from below the surface soil horizons at available sites within the city to allow comparison between lead levels in the urban environment, and those levels occurring naturally in the soil (background).

Approximately 500 g of soil was collected with a plastic trowel and deposited into a paper sample bag. The trowel was rinsed with water and wiped off between each sample. Effort was made to collect exposed surface soil, mainly from the top 2 cm, but samples were taken at greater depths (up to 5 cm.) due to the presence of ground cover at some sites. Duplicate samples were collected approximately every tenth site.

Soil samples were analysed in the Geological Survey's Geochemical Laboratory in St. John's. The soil was air-dried in ovens at 40°C and dry-sieved through 63 μm stainless steel sieves before geochemical analysis. Samples were digested with a strong acid to ensure complete dissolution. This was true for most elements, with the exceptions being Cr from chromite, Ba from barite and Zr from zircon as these minerals are not usually completely dissolved. Inductively coupled plasma emission spectrometry (ICP) was then used to analyse for trace elements.

In addition to lead, the following elements were determined: aluminium, barium, beryllium, calcium, cerium, cobalt, chromium, copper, dysprosium, iron, gallium, potassium, lanthanum, lithium, magnesium, manganese, molybdenum, sodium, niobium, nickel, phosphorus, scandium, strontium, titanium, vanadium, yttrium, zinc and zirconium. Data quality was monitored using laboratory duplicates (analytical decision only). Accuracy estimates are provided by the results from standard reference materials analysed with them.

Statistical Analysis

Discriminate analysis is an exploratory statistical procedure that determines how well it is possible to separate two or more groups based on the values of several variables for those groups (Manly1994). It uses Mahalanobis distances to calculate how far individual cases are from the group means for the variables, and assigns the case to the group with the closest mean.

Using the large set of metal data collected through ICP-MS analysis of the soil samples it is possible to see if certain metals are more characteristic of different sources of pollution by using discriminant analysis. If this occursr then the predicted major source of contamination for each soil sample can be confirmed, and future soil samples can be entered into the discriminant analysis equation to determine the source of pollution.

Before the statistical analysis was performed the soil samples were classified into source groups based on measured distances from roadways and buildings. Sources labelled paint were located less than 2 meters from a building. Sources labelled road were located less than four meters from a road, while backyard samples from houses downtown were labelled coal sources, as coal pieces were observed at several of these locations. In addition samples that were well away from buildings, roads, and not in older backyards were labelled ambient samples as they were representative of background urban pollution. There were also several samples taken at depth from the St. John's area that pre-date urbanization. These samples were labelled as background and were assumed to represent the natural metal content of the local soil due to the weathering of bedrock. Some samples were difficult to label as they seemed to overlap several groups. These samples were labelled as a combination of sources but were left out of the discriminant analysis as they may have diluted the source-specific results and caused misclassification errors.

Discriminate analysis is based on two assumptions; that the within-group covariance matrix is the same for all groups, and that the data is normally distributed between groups (Manly 1994). If the first assumption is not met it is possible to use quadratic discriminant analysis, which does not assume that the groups have equal covariance matrices. In quadratic analysis the observation is still classified in the group that has the smallest squared distance, but that distance no longer simplifies into a linear function (Minitab 14 2004). Even though the within-group covariance matrices were not the same for all groups (see Appendix 1), a linear discriminant analysis was still used, as Minitab would not allow a quadratic analysis due to the fact that arsenic was too highly correlated with other predictors in the background group.

There are several other options to decide upon before running a discriminant analysis. First it is possible to select *a priori* probabilities for classifying cases into groups. This is done if the probability of membership is inherently different between groups, and if that probability is known (Manly 1994). This option will not be used for our dataset as there is an equal probability that the contamination for a sample will be from any one source.

Secondly, it is possible to run a step-wise discriminant analysis. This procedure adds one variable to the function at a time in order to see when adding further variables no longer improves classification (Manly 1994). This approach will also not be used in the analysis because as this point we have no reason to believe that some metals will explain more of the source differences than others. We also do not want to lose any information about the metal concentrations by eliminating some from the analysis right away.

Results

Discriminant Analysis: Pollution Source versus Metal Concentration

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Linear Method for Response: Source


Predictors: Mo, Cr, P, Zn, Pb, Co, Ni, Fe, Mg, Cd, Ti, V, Be, Ca, Nb, Cu, Na,
            Zr, Dy, Sc, Y, Al, Mn, Sr, La, Ce, Ba, Li, K, Ars


Group          A          B          C          P          R
Count        165          7         34         32         83
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Table 1: Summary of Classification of Soil Samples into Pollution Source Groups

	True Group				
Put into Group	A	B	C	P	R
A	128	1	1	6	19
B	4	6	1	0	2
C	3	0	27	7	1
P	7	0	4	14	3
R	23	0	1	5	58
Total N	165	7	34	32	83
N correct	128	6	27	14	58
Proportion	0.776	0.857	0.794	0.438	0.699

N = 321 N Correct = 233 Proportion Correct = 0.726

Overall the discriminant function had a 72.6% success rate in classifying the samples according to potential source of metal contamination (Table 1). The background samples were predicted the best, while paint samples were predicted the worst, often being mistaken for coal samples. The three other source types were predicted with the same success frequency. Ambient and road samples were often misclassified as each other, background were frequently mistaken for ambient samples, and coal samples were most often misclassified as paint samples.

These relationships are seen again when examining the squared distance between groups. Groups with means that are close together in discriminant analysis are most similar, therefore according to the results in Table 2, ambient and road samples have the most similar metal composition with a squared distance of 2.4511. Coal and paint samples are also fairly close together (5.3927), but the background samples remain fairly distinct from all groups, especially the coal and paint samples (30.2865 and 28.1502).

Table 2: Squared Distance Between Pollution Source Groups

	A	B	C	P	R
A	0.0000	18.1677	10.6024	6.5666	2.4511
B	18.1677	0.0000	30.2865	28.1502	18.8280
C	10.6024	30.2865	0.0000	5.3927	11.9689
P	6.5666	28.1502	5.3927	0.0000	6.6237
R	2.4511	18.8280	11.9689	6.6237	0.0000

The 73% overall classification success rate may be overly optimistic because the data being classified were also used to create the classification function (Minitab, 2004). In order to account for this potential error cross-validation should be used to generate the classification. In this statistical procedure the classification function is recalculated repeatedly by omitting one observation at a time, and then classifying that omitted observation using the new value for the classification function (Minitab 14 2004). The results of the cross-validation classification are presented in Table 3.

Table 3: Summary of Classification with Cross-validation of Soil Samples into Pollution Source Groups

	True Group				
Put into Group	A	B	C	P	R
A	115	3	2	8	21
B	7	4	1	0	2
C	4	0	25	9	1
P	8	0	5	9	6
R	31	0	1	6	53
Total N	165	7	34	32	83
N correct	115	4	25	9	53
Proportion	0.697	0.571	0.735	0.281	0.639

N = 321 N Correct = 206 Proportion Correct = 0.642

An examination of the cross validation classification table for the entire dataset shows that all groups were classified less successfully. However the group that suffered the greatest reduction in successful classification were the background samples, from 86% to 57%. This may have been due to low sample size (n=7), as not including one value in the group mean calculation could significantly alter the result and effect the classification. The paint group also saw a dramatic decrease in classification success to only 28%.

The linear discriminant function coefficients are displayed in Table 4. The discriminant analysis equation takes the form:

$$\text{Classification Score} = \beta_0 + \beta_{Mo}*Mo + \beta_{Cr}*Cr + \beta_P*P + \ldots$$

Where β_0 is the constant and β_X*X represents the product of the metal specific coefficient and the concentration of that metal.

Table 4: Linear Discriminant Function for Pollution Source Groups

	A	B	C	P	R
Constant	-128.92	-137.19	-141.03	-141.07	-128.89
Mo	-0.69	-2.01*	-0.41	-0.63	-0.42
Cr	0.06	0.07	0.06	0.07	0.09**
P	0.01	0.01	0.01	0.01	0.01
Zn	-0.01	-0.02	-0.01	-0.01	-0.02
Pb	-0.01	-0.01	-0.01	-0.01	-0.01
Co	0.19	-0.25*	0.48**	0.44**	0.28
Ni	-0.18	0.01**	-0.10	-0.21	-0.14
Fe	6.43	7.90**	5.47	6.20	5.98
Mg	-63.66	-88.41*	61.04	-61.05	-64.06
Cd	-1.65	-2.01	-2.97*	-1.95	-0.95
Ti	0.01	0.01	0.01	0.01	0.01
V	0.29**	0.19	0.23	0.31**	0.30**
Be	-14.99	-14.73	-17.17	-19.37*	-18.34
Ca	24.74	29.75**	24.94	24.85	25.89
Nb	1.12	1.14	1.29**	1.09	1.10
Cu	-0.01	-0.00	-0.02	-0.00	-0.00
Na	4.48	8.17**	6.82	5.03	7.43
Zr	0.21	0.22	0.23	0.22	0.21
Dy	4.49	21.19**	5.73	4.04	5.83
Sc	-0.45	2.04**	-0.45	-1.74	-0.72
Y	-0.76	-4.42*	-0.83	-0.78	-0.91
Al	3.82	4.79**	3.41	3.98	3.71
Mn	0.01	0.01	0.01	0.01	0.01
Sr	0.11	0.10	0.13	0.14**	0.10
La	2.89	2.39	2.99	3.26**	2.78
Ce	-1.31	-0.84	-1.45*	-1.41*	-1.32
Ba	0.01	0.01	0.01	0.01	0.01
Li	0.59	0.48	0.67	0.76**	0.76**
K	50.03	49.50	48.49	49.27	50.92
Ars	-0.05*	-0.02	-0.02	-0.05*	-0.06*

* high absolute values for negative coefficients
** high absolute values for positive coefficients

Because these coefficients are dependent on the concentration of metal present it is difficult to read down the column and see which metals contribute the most to the discriminant function equation for each source. A better approach would be to find the standardized coefficients, but this was not possible in Minitab. However some patterns do emerge by examining the rows of coefficients which are comparable because they represent the same metal.

The ambient samples did not display extreme coefficients for any of the variables. However the discriminant function did have a somewhat high positive coefficient for vanadium and negative coefficient for arsenic, but these extreme values were also shared by the paint and road groups.

The discriminant function for background samples was the most unique with the lowest negative coefficients for molybdenum, cobalt, magnesium, and yttrium, and the highest positive coefficients for nickel, iron, calcium, sodium, dysprosium, scandium, and aluminum.

High positive coefficients for the coal group were found for cobalt and niobium. Negative coefficients for this group included cadmium and cerium. It is interesting to note that the coal group discriminant function coefficients shared many characteristics with the paint group.

The paint group showed similarities with both the coal and the road groups. High positive coefficients were associated with cobalt, vanadium, strontium, lanthanum, and lithium, while beryllium, cerium, and arsenic had more extreme negative coefficients.

Lastly the road group had high positive coefficients for chromium, vanadium and lithium, and a high negative coefficient for arsenic. The discriminant function for this group showed similarities with both the paint and ambient groups.

Some of the coefficients of the metals analyzed did not show significant variation between groups. They included phosphorous, zinc, lead, titanium, copper, zirconium, manganese, barium, and potassium. The values for these metals must not have been very different between the source groups, either being consistently low from natural background sources (ie. bedrock weathering), or consistently high due to widespread anthropogenic pollution. Alternatively there may have been too much intra-group variation to make a clear relationship between metal concentration and group membership.

In order to see if I could improve the classification process I ran three more discriminant analyses.

Discriminant Analysis: Expanded Pollution Sources versus Metal Concentration

In the second discriminant analysis I added another group characteristic, location within the city, either downtown (D) or suburban (S). This created a total of eight categories for source group: AS, AD, BS, CD, PS, PD, RS, RD. I added this variable to the analysis because I felt that samples taken from older, more central parts of the city might have a significantly different composition than similar samples taken in the newer suburban districts due to the fact that older properties would have had a longer history of anthropogenic contamination.

```
Linear Method for Response: Expanded Source


Predictors: Mo, Cr, P, Zn, Pb, Co, Ni, Fe, Mg, Cd, Ti, V, Be, Ca, Nb, Cu, Na,
            Zr, Dy, Sc, Y, Al, Mn, Sr, La, Ce, Ba, Li, K, Ars
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Group	AD	AS	BS	CD	PD	PS	RD	RS
Count	69	96	7	34	25	7	67	16

Table 5: Summary of classification of Soil Samples into Expanded Pollution Source Groups

	True Group							
Put into Group	AD	AS	BS	CD	PD	PS	RD	RS
AD	40	11	0	4	2	1	15	2
AS	11	69	1	0	0	1	3	4
BS	0	4	6	1	0	0	1	0
CD	1	1	0	27	5	0	1	0
PD	1	0	0	2	12	0	1	0
PS	4	4	0	0	2	4	4	0
RD	12	3	0	0	4	0	36	3
RS	0	4	0	0	0	1	6	7
Total N	69	96	7	34	25	7	67	16
N correct	40	69	6	27	12	4	36	7
Proportion	0.580	0.719	0.857	0.794	0.480	0.571	0.537	0.438

N = 321 N Correct = 201 Proportion Correct = 0.626

Table 6: Summary of Classification with Cross-validation of Samples into Expanded Pollution Source Groups

Put into Group	AD	AS	BS	CD	PD	PS	RD	RS
	True Group							
AD	32	12	0	4	2	1	16	4
AS	11	57	2	0	0	3	4	4
BS	1	5	4	1	0	0	1	0
CD	2	2	0	22	10	0	1	0
PD	1	1	0	5	7	0	2	0
PS	5	8	0	1	2	1	4	1
RD	16	3	1	0	3	1	32	4
RS	1	8	0	1	1	1	7	3
Total N	69	96	7	34	25	7	67	16
N correct	32	57	4	22	7	1	32	3
Proportion	0.464	0.594	0.571	0.647	0.280	0.143	0.478	0.188

N = 321 N Correct = 158 Proportion Correct = 0.492

Successful classification was not improved by adding the additional downtown/suburban group (Tables 5 and 6). In particular the discriminant analysis function found it difficult to classify downtown paint and suburban paint and road samples.

Table 7 describes the similarities between groups as a function of distance between group means. It shows that in suburban areas paint and background samples are similar to ambient samples, though paint and background samples themselves may not be that alike. The table also points out that road samples are similar regardless of whether they occur downtown or in the suburbs, and that in the downtown area the composition of paint and coal samples are similar.

Table 7: Squared Distance Between Expanded Pollution Source Groups

	AD	AS	BS	CD	PD	PS	RD	RS
AD	0.0000	2.8438	20.1919	10.2922	8.7927	7.6021	1.8923	4.7483
AS	2.8438	0.0000	17.8880	14.4861	14.4793	6.6751	4.9905	5.4694
BS	20.1919	17.8880	0.0000	32.0016	33.3319	27.8156	19.0750	23.4207
CD	10.2922	14.4861	32.0016	0.0000	5.1812	18.5929	12.4176	16.3474
PD	8.7927	14.4793	33.3319	5.1812	0.0000	15.7923	9.4333	14.5305
PS	7.6021	6.6751	27.8156	18.5929	15.7923	0.0000	9.1954	11.2596
RD	1.8923	4.9905	19.0750	12.4176	9.4333	9.1954	0.0000	3.8162
RS	4.7483	5.4694	23.4207	16.3474	14.5305	11.2596	3.8162	0.0000

The coefficient trends for the second discriminant analysis (Table 8) are very similar to the first analysis, and also follow the similarities between sources discussed in the previous paragraph.

Table 8: Linear Discriminant Function for Expanded Pollution Source Groups

	AD	AS	BS	CD	PD	PS	RD
Constant	-130.32	-127.65	-136.63	-141.00	-141.64	-145.18	-129.45
Mo	-0.51	-0.77	-2.02*	-0.36	-0.58	-0.56	-0.45
Cr	0.07	0.06	0.07	0.06	0.07	0.07	0.08
P	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Zn	-0.01	-0.01	-0.02	-0.01	-0.01	-0.01	-0.02
Pb	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01
Co	0.29	0.21	-0.19	0.52	0.43	0.69**	0.36
Ni	-0.08	-0.21*	0.03	-0.05	-0.14	-0.19*	-0.08
Fe	5.68	6.73	7.85**	4.96	5.37	7.00**	5.70
Mg	-64.49	-64.67	-89.34*	-60.58	-58.25	-70.98	-65.44
Cd	-0.79	-1.94	-1.87	-2.29	-0.86	-2.98*	-0.37
Ti	0.01	0.01	0.01	0.01	0.01	0.02	0.01
V	0.32	0.28	0.20	0.24	0.33	0.31	0.31
Be	-17.91	-14.53	-15.61	-19.01	-21.56*	-20.05	-20.61
Ca	24.75	24.78	29.78**	24.76	24.33	26.23	26.04
Nb	0.84	1.08	1.00	1.15**	1.04	0.54	0.88
Cu	-0.01	-0.01	-0.01	-0.02	-0.01	-0.01	-0.01
Na	5.18	4.10	7.95	6.84	4.83	6.00	7.04
Zr	0.19	0.21	0.21	0.21	0.18	0.25	0.18
Dy	7.48	3.64	21.50**	7.54	6.46	3.59	7.43
Sc	-1.42	-0.08	1.95**	-1.05	-2.61	-1.17	-1.14
Y	-0.98	-0.73	-4.45*	-0.97	-0.94	-0.86	-1.07
Al	3.43	3.94	4.73*	3.16	3.59	4.25*	3.55
Mn	0.01	0.01	0.01	0.01	0.01	0.00	0.01
Sr	0.09	0.11	0.10	0.13	0.15**	0.10	0.09
La	2.78	2.90	2.38	2.93	3.17**	3.27**	2.76
Ce	-1.35	-1.27	-0.83	-1.47*	-1.45*	-1.35	-1.32
Ba	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Li	0.80	0.50	0.49	0.82	1.01**	0.51	0.87
K	54.52**	49.20	50.70	50.99	51.86	51.99	54.15**
Ars	-0.00	-0.05	-0.00	0.01	-0.02	-0.02	-0.03

	RS
Constant	-127.36
Mo	0.02**
Cr	0.09**
P	0.01
Zn	-0.02
Pb	-0.01
Co	0.25
Ni	-0.12
Fe	5.48
Mg	-61.66
Cd	-1.32
Ti	0.01
V	0.29
Be	-16.37
Ca	25.15
Nb	1.16**
Cu	-0.00
Na	9.82**
Zr	0.28**
Dy	6.13
Sc	-1.07
Y	-0.80
Al	3.49
Mn	0.01
Sr	0.08
La	2.61
Ce	-1.38

Ba	0.01
Li	0.74
K	48.59
Ars	-0.07*

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* high absolute values for negative coefficients
** high absolute values for positive coefficients
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The ambient samples once again did not have any extreme coefficients. The downtown ambient group only shared a high positive coefficient for potassium with the downtown road group, and the suburban ambient group had a high negative coefficient for nickel similar to the suburban paint sample.

The discriminant function for background samples was the most unique with the lowest negative coefficients for chromium, magnesium, yttrium, and aluminum, and the highest positive coefficients for iron, calcium, dysprosium, and scandium.

High positive coefficients for the coal group were found for niobium, and negative coefficients for this group included cerium.

Both paint groups had high positive coefficients for lanthanum, but the downtown group also had high strontium and lithium coefficients, while cobalt and iron were more important for the suburban group. Beryllium and cerium were metals with more extreme negative coefficients for the downtown paint group, and their counterparts for the suburban paint group were nickel, cadmium, and aluminum

Lastly the two road groups had fairly different coefficients. The downtown road group had high positive coefficients for potassium, while the suburban road group had molybdenum, chromium, niobium, sodium, and zirconium. The suburban group also had a high negative coefficient for arsenic.

There are several reasons why this discriminant analysis might have been unsuccessful. First there might not have been enough variation in metal concentrations between groups to distinguish one source from the other. Problems might have also occurred solely because the large number of groups created increased the chance of misclassification. To investigate the former problem I separated the data set into downtown and suburban samples and ran discriminant analysis on each separately.

Discriminant Analysis: Downtown Source versus Metal Concentration

```
Linear Method for Response: Downtown Source


Predictors: Mo, Cr, P, Zn, Pb, Co, Ni, Fe, Mg, Cd, Ti, V, Be, Ca, Nb, Cu, Na,
            Zr, Dy, Sc, Y, Al, Mn, Sr, La, Ce, Ba, Li, K, Ars
```

Group	A	C	P	R
Count	69	34	25	67

Note that there were no background samples taken in downtown St. John's.

Table 9: Summary of Classification of Soil Samples into Downtown Pollution Source Groups

	True Group			
Put into Group	A	C	P	R
A	52	2	4	10
C	1	27	4	0
P	3	4	13	2
R	13	1	4	55
Total N	69	34	25	67
N correct	52	27	13	55
Proportion	0.754	0.794	0.520	0.821

```
N = 195           N Correct = 147          Proportion Correct = 0.754
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Table 10: Summary of Classification with Cross-validation for Downtown Pollution Source Groups

	True Group			
Put into Group	A	C	P	R
A	42	4	4	18
C	2	23	6	1
P	5	6	10	5
R	20	1	5	43
Total N	69	34	25	67
N correct	42	23	10	43
Proportion	0.609	0.676	0.400	0.642

```
N = 195           N Correct = 118          Proportion Correct = 0.605
```

Classification of downtown samples into pollution source groups was roughly as successful as the classification of the entire database into the original five groups. It was a little better in the classification and a little worse in the cross-validation. This may be due to a smaller sample size. Paint once again is the hardest to classify.

The distances between group means (Table 11) followed the same pattern and almost the same magnitude as the first discriminant analysis. This suggests that there was not a lot of improvement by separating the samples into location.

Table 11: Squared Distance Between Downtown Pollution Source Groups

	A	C	P	R
A	0.0000	8.5996	7.1661	2.9594
C	8.5996	0.0000	5.0696	11.7980
P	7.1661	5.0696	0.0000	8.3490
R	2.9594	11.7980	8.3490	0.0000

It is difficult to discuss the coefficients for the downtown pollution sources (Table 12) because there is no background group included in the analysis. This omission greatly changes the values of the coefficients and makes a comparison with the original discrimination difficult.

Table 12: Linear Discriminant Function for Downtown Pollution Source Groups

	A	C	P	R
Constant	-160.43	-171.63	-167.57	-162.35
Mo	-0.84	-0.32	-0.97	-0.91
Cr	0.12	0.11	0.12	0.14
P	0.01	0.01	0.01	0.01
Zn	-0.04	-0.04	-0.03	-0.04
Pb	-0.01	-0.01	-0.01	-0.01
Co	1.38	1.37	1.41	1.56
Ni	-0.31	-0.21	-0.31	-0.26
Fe	4.43	3.04	3.94	3.86
Mg	-5.31	4.45	4.89	-11.38
Cd	1.64	0.25	1.30	1.80
Ti	0.02	0.02	0.02	0.02
V	0.41	0.34	0.41	0.40
Be	3.22	4.45	0.84	0.00
Ca	25.18	25.49	24.15	26.49
Nb	1.15	1.37	1.44	1.06
Cu	-0.09	-0.10	-0.08	-0.08
Na	22.90	26.83	22.27	29.11
Zr	0.42	0.43	0.35	0.39
Dy	-20.74	-20.46	-22.49	-22.82
Sc	0.14	0.46	-0.90	0.61
Y	4.99	4.87	5.08	5.06
Al	1.35	1.06	1.24	0.76
Mn	-0.00	0.00	0.00	-0.00
Sr	-0.01	-0.00	0.04	-0.02
La	0.81	0.93	1.00	0.43
Ce	-1.31	-1.44	-1.37	-1.16
Ba	0.01	0.01	0.01	0.00
Li	-0.84	-0.88	-0.60	-0.58
K	48.87	43.22	46.84	50.43
Ars	-0.02	-0.01	-0.05	-0.04

Discriminant Analysis: Suburban Pollution Source versus Metal Concentration

```
Linear Method for Response: Suburban Pollution Source


Predictors: Mo, Cr, P, Zn, Pb, Co, Ni, Fe, Mg, Cd, Ti, V, Be, Ca, Nb, Cu, Na,
            Zr, Dy, Sc, Y, Al, Mn, Sr, La, Ce, Ba, Li, K, Ars
```

Group	A	B	P	R
Count	96	7	7	16

Table 13: Summary of Classification of Soil Samples into Suburban Pollution Source Groups

	True Group			
Put into Group	A	B	P	R
A	86	1	1	5
B	3	6	0	0
P	3	0	6	0
R	4	0	0	11
Total N	96	7	7	16
N correct	86	6	6	11
Proportion	0.896	0.857	0.857	0.688

N = 126 N Correct = 109 Proportion Correct = 0.865

Table 14: Summary of Classification with Cross-validation of Soil Samples into Suburban Pollution Source Groups

	True Group			
Put into Group	A	B	P	R
A	73	3	4	6
B	9	4	0	0
P	7	0	3	1
R	7	0	0	9
Total N	96	7	7	16
N correct	73	4	3	9
Proportion	0.760	0.571	0.429	0.563

N = 126 N Correct = 89 Proportion Correct = 0.706

The classification of suburban samples according to pollution source was much better than the original discriminant analysis. In the analysis the road samples were the problem, with several being misclassified as ambient samples. However this changed in the cross-validation where paint samples once again appeared to be difficult to identify. In this case they were mistaken for ambient samples.

The distances between suburban group means (Table 15) were greater than in the original discriminant analysis indicating better separation between groups.

Table 15: Squared Distance Between Suburban Pollution Source Groups

	A	B	P	R
A	0.0000	16.2599	13.2036	9.8127
B	16.2599	0.0000	35.4339	27.0595
P	13.2036	35.4339	0.0000	18.1435
R	9.8127	27.0595	18.1435	0.0000

Once again it is difficult to discuss the coefficients for this subset of data (Table 16) because not all source groups are included in the analysis. In this case is no coal group in the suburbs. This omission greatly changes the values of the coefficients and makes a comparison with the original discriminant analysis difficult.

Table 16: Linear Discriminant Function for Suburban Pollution Source Groups

	A	B	P	R
Constant	-198.98	-216.84	-224.59	-194.08
Mo	-0.42	-1.62	0.00	0.40
Cr	-0.34	-0.33	-0.33	-0.19
P	0.01	0.02	0.01	0.01
Zn	-0.01	-0.00	-0.02	-0.02
Pb	-0.00	-0.01	0.02	0.00
Co	-0.78	-0.95	-0.01	-0.65
Ni	0.16	0.48	0.17	0.32
Fe	1.63	5.19	1.81	-0.37
Mg	-167.35	-187.32	-200.96	-169.47
Cd	16.64	18.83	12.55	13.23
Ti	0.01	0.01	0.02	0.01
V	0.99	0.88	1.07	1.11
Be	-27.16	-33.58	-37.55	-34.05
Ca	41.90	41.03	52.71	42.05
Nb	0.39	0.39	-0.39	0.81
Cu	0.01	-0.02	0.00	-0.01
Na	7.31	9.94	7.19	12.19
Zr	0.56	0.59	0.63	0.57
Dy	32.99	47.02	32.93	35.06
Sc	-1.87	-0.57	-3.39	-3.30
Y	-6.89	-9.02	-7.89	-6.31
Al	8.14	9.07	10.08	8.51
Mn	0.02	0.02	0.02	0.02
Sr	0.41	0.43	0.43	0.41
La	4.62	3.99	5.97	4.30
Ce	-1.90	-1.40	-2.11	-1.97
Ba	0.01	0.02	-0.03	0.00
Li	2.35	2.18	2.30	2.54
K	76.38	74.52	89.85	73.18
Ars	-0.05	-0.04	-0.02	-0.04

Discussion

Prediction of Pollution Source

Separating the data by location before running the discriminant analysis appears to be the most successful method of predicting the pollution source for soil samples in St. John's.

While it does not improve classification for samples downtown it greatly raises the success rate for suburban samples.

The fact that the classification of downtown samples did not improve after separation from suburban samples indicates that there is not enough variation in metal concentration between groups to distinguish between them. It could be that the pollution in the downtown core of St. John's is so uniform that attempting to assign one source is not possible. This is a plausible explanation as the physical distances between painted houses, roadways, and coal disposal in backyards are not very great.

Similarities Between Sources

Figures 1 and 2 show some interesting relationships between potential pollution sources based on the Mahalanobis distances between group means. Those groups with means close together are assumed to be similar in metal content. Similarities are represented in the figures by arrows, with the direction of the arrow indicating which group has similar characteristics to another group.

Figure 1 represents pollution sources for the whole city and shows two distinct groups, paint and coal, and ambient, road, and background samples. Paint and coal sources may be similar because they are both taken from residential properties. It is possible that weathered paint may have been transferred to the entire backyard of older properties influencing the metal composition. Paint and coal may also just have similar metal constituents to begin with.

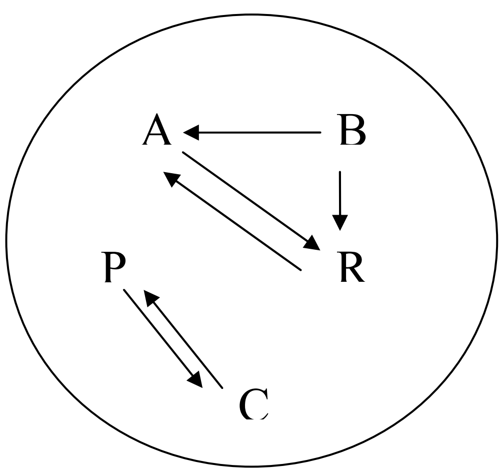

Figure 1: Similarities between Pollution Sources based on Squared Distances between Means

Figure 2 represents the city divided into the downtown core and the suburban areas. The Mahalanobis distances for this analysis show that there are many similarities between the within and between the two spatial divisions.

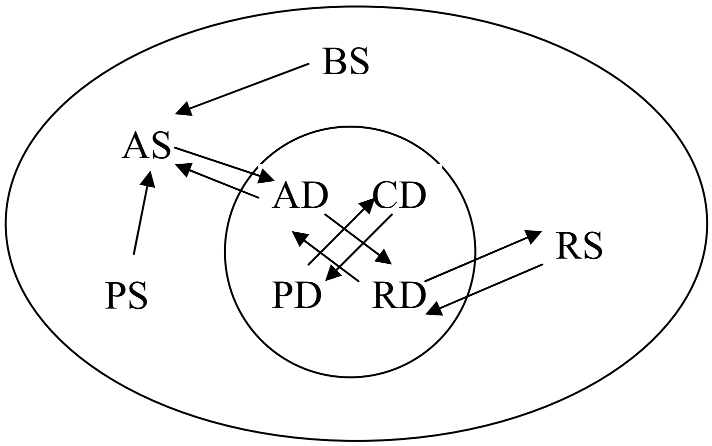

Figure 2: Similarities Between Pollution Sources Considering the Location within the City

Downtown, ambient and road samples share many characteristics. This may be because the pollution associated with gasoline combustion can spread over a large area, affecting not only the samples in close proximity to roads, but also samples taken at further distances. It is also possible that the cut-off distance used when classifying samples into road and ambient sources was too small. As I have mentioned in the methodology section, samples closer than 4 meters to a street were defined as having vehicle emissions as the major source of metals, but perhaps this distance is not representative of the true impact that roads have. It may be necessary to go back and reclassify the samples to include all samples taken say within 10 meters of the road, and then run the discriminant analysis again to see whether the Mahalanobis distances between ambient and road sources increase.

Coal and paint samples are also similar in the downtown core. As mentioned with Figure 1 this may be due to the close proximity of paint and coal samples. Often they are both taken from the backyard and both sources of metal contamination may be present in both samples.

Within the suburban group, ambient, background and paint samples seem to group together. This makes sense as newer areas of the city have had less contaminant input to the soil, making surface (ambient) and sub-surface (background) samples very similar. While it might at first seem odd that samples labelled as having paint as the major source of pollution are also in this group, it is important to remember that the metal content of paint, especially for lead, has been regulated for the last thirty years. Many of the houses in the suburban districts would have never been covered with leaded paint and therefore the metal signature for this source would not be very different from ambient of background samples.

There are also several similarities between the two spatial groups. Location in either the downtown core or in the suburbs does not seem to influence the metal composition of road samples. This may be because although residential development may be new to the suburban districts the roads running through them may be quite old and therefore may have had the same opportunity to accumulate metals in the surrounding soil.

Ambient sources also show this lack of distinction between downtown and suburban locations. This is a pattern that is more difficult to explain. Perhaps further research may uncover reasons for these similarities.

Relationships Between Sources and Metals

As mentioned earlier in the paper each of the three sources; paint, vehicle emissions, and coal combustion could have an association with a particular set of metals. And this association may be important in classifying the source of metal contamination. However the metals deemed important to the discriminant analysis function for each of the sources were not similar to the metals listed as common for those sources in the literature. Perhaps comparison between the two is not possible. The discriminant function looked for metals that distinguished the pollution source from all other groups, not necessarily for metals with the highest concentrations as certain metals might be high in all source groups and therefore not good classification predictors. For example the literature mentions zinc and lead as present in all pollution sources tested in this analysis, however neither of these metals received large absolute coefficient values.

In order to determine if the concentration of each metal is associated with the pollution source it may be more beneficial to run a multiple analysis of variance, using metal concentration as the response variable and pollution source as a categorical explanatory variable.

Conclusions

Discriminant analysis of heavy metal concentrations in the soil of St. John's provided only a moderately successful method for classifying soil samples according to pollution source. The best model ran the analysis after separating by location and had a cross-validation classification success of 61% for samples collected downtown and a 71% success rate for suburban samples.

Now that patterns in the data are better understood future analysis of this dataset should focus on confirmatory analyses to determine if significant differences in metal concentrations occur between sources of metal contamination.

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Appendix 1

Covariance Matrices for Pollution Source Groups

Pooled Covariance Matrix

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V	Be
Mo	8												
Cr	21	1067											
P	584	6203	872962										
Zn	177	3537	104273	53183									
Pb	402	6250	230107	82226	304123								
Co	7	35	1364	362	781	22							
Ni	29	150	3478	1118	2897	38	148						
Fe	4	14	566	100	270	5	17						
Mg	-0	0	-12	0	-2	0	0						
Cd	0	4	211	91	170	1	2						
Ti	-304	-1147	39912	-20911	-7288	707	-323						
V	17	208	8897	1654	5951	47	134						
Be	1	3	118	36	83	2	4						
Ca	0	1	61	28	34	0	1						
Nb	0	-5	383	-16	92	3	3						
Cu	300	975	12053	6046	11589	262	1237						
Na	-0	1	-142	17	10	-0	-1						
Zr	-5	5	-1362	-562	-1126	16	-7						
Dy	0	-1	-99	-2	2	2	2						
Sc	0	2	131	-21	-20	4	3						
Y	3	2	-449	36	58	10	14						
Al	-0	-1	-108	-20	-41	0	-1						
Mn	80	1093	24856	7978	12249	1130	659						
Sr	37	77	-5441	953	2054	15	153						
La	3	2	-803	71	111	12	17						
Ce	5	4	-1138	118	155	27	34						
Ba	126	1118	19947	20643	52449	67	595						
Li	10	21	2829	268	841	30	55						
K	-0	-1	-157	-7	-25	-0	-1						
Ars	77	174	7730	1420	3612	79	290						

Fe	3						
Mg	-0	0					
Cd	0	0	0				
Ti	115	37	-45	503701			
V	16	1	3	6166	444		
Be	1	0	0	-2	3	0	
Ca	-0	0	0	-84	-1	0	0
Nb	1	0	-0	1265	18	0	-0
Cu	161	-2	9	-13766	550	33	5
Na	-0	0	0	-34	-3	-0	0
Zr	2	1	-2	6208	67	1	-2
Dy	0	0	0	220	3	0	-0
Sc	1	0	-0	955	19	0	-0
Y	2	0	0	982	14	1	-0
Al	-0	0	-0	159	1	0	-0
Mn	120	13	27	38710	1186	60	-12
Sr	11	-0	2	-6231	-126	4	6
La	1	0	0	297	12	2	-0
Ce	4	1	0	2314	40	3	-1
Ba	26	-3	50	-50387	-82	18	22
Li	10	0	0	3605	113	3	-1
K	-0	0	-0	-61	-2	-0	-0
Ars	45	-1	1	-2180	178	10	1

Nb	7						
Cu	-10	15052					
Na	-0	-7	0				
Zr	23	-183	-1	278			
Dy	1	12	0	5	1		
Sc	3	-3	-0	16	1	3	
Y	5	95	0	26	3	4	17
Al	0	-13	0	4	0	0	0
Mn	187	2411	-18	1117	118	234	691
Sr	-29	1695	6	-181	-1	-14	8
La	3	124	0	21	4	4	20
Ce	14	208	0	67	7	10	37
Ba	-148	5589	32	-922	-21	-125	-52
Li	16	320	-2	75	4	13	22
K	-0	-8	0	-0	0	-0	0
Ars	8	2938	-3	-49	3	4	25

Al	1						
Mn	-5	173508					
Sr	-1	-2003	1018				
La	0	859	15	30			
Ce	1	1819	3	47	103		
Ba	-11	-3999	2792	31	-142	46045	
Li	1	1236	-67	20	57	-505	90
K	0	-7	3	0	0	19	-1
Ars	-3	422	422	32	63	870	123

K	0	
Ars	-4	973

Covariance matrix for Group A

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V	Be
Mo	12												
Cr	16	166											
P	70	3842	649250										
Zn	38	799	43737	28324									
Pb	108	1599	85193	24409	71284								
Co	10	40	699	230	368	27							
Ni	44	103	1484	407	1022	50	197						
Fe	7	13	323	28	119	7	26						
Mg	-0	1	10	3	5	0	0						
Cd	-0	1	91	35	54	0	0						
Ti	-372	3091	203500	6242	19143	1352	332						
V	18	180	9051	948	2171	57	139						
Be	1	3	4	15	29	2	5						
Ca	0	0	29	13	12	0	0						
Nb	1	6	597	21	120	4	5						
Cu	508	709	-1351	2194	4354	426	1996						
Na	-1	-2	-185	6	-7	-1	-2						
Zr	-8	44	1252	-37	-159	33	4						
Dy	1	3	-54	14	31	3	4						
Sc	0	12	477	36	67	7	7						
Y	5	15	-385	84	146	16	24						
Al	-1	-1	-110	-15	-45	0	-2						
Mn	128	2046	5336	11888	6500	1569	1033						
Sr	60	-24	-9902	-239	-109	12	188						
La	5	14	-1252	74	84	17	27						
Ce	10	37	-1159	244	449	40	57						
Ba	76	-50	-47906	3697	12587	-114	122						
Li	16	72	3410	358	760	44	85						
K	-0	-1	-177	-6	-20	-1	-2						

Ars	129	188	1755	177	1166	106	444

Fe	4						
Mg	-0	0					
Cd	-0	0	0				
Ti	234	61	-10	623157			
V	20	1	1	9592	449		
Be	1	0	0	-18	3	0	
Ca	-0	0	0	-45	-1	0	0
Nb	2	0	-0	1562	25	0	-0
Cu	274	-3	-3	-17106	736	53	5
Na	-0	0	0	-101	-5	-0	0
Zr	2	1	-1	7982	128	1	-1
Dy	0	0	0	233	5	0	0
Sc	1	0	0	1286	28	0	-0
Y	3	0	0	994	23	2	0
Al	-0	0	-0	145	-1	-0	-0
Mn	172	23	44	53233	1470	87	0
Sr	20	-1	0	-10301	-231	5	4
La	2	0	0	-302	10	2	0
Ce	7	1	0	2141	60	4	-0
Ba	-20	-5	12	-70372	-1345	9	13
Li	14	1	0	5503	158	3	-1
K	-0	0	-0	-134	-4	-0	0
Ars	71	-1	-2	-2047	235	14	0

Nb	8						
Cu	-3	24482					
Na	-0	-18	0				
Zr	26	-354	-2	282			
Dy	1	24	-0	6	1		
Sc	3	9	-0	22	1	4	
Y	4	173	-0	28	4	5	22
Al	0	-21	0	4	-0	0	-0
Mn	180	4309	-24	1631	179	349	1097
Sr	-37	2841	7	-259	-1	-25	5
La	2	223	0	20	5	4	28
Ce	13	402	-0	76	9	13	48
Ba	-192	4236	42	-1075	-27	-169	-84
Li	20	609	-3	106	6	19	29
K	-0	-18	0	-1	-0	-0	-0
Ars	10	5209	-5	-57	7	8	50

Al	1						
Mn	-6	273460					
Sr	-0	-2056	1102				
La	-0	1367	21	44			
Ce	0	2617	10	64	127		
Ba	2	-8650	3256	88	-112	24339	
Li	1	1778	-91	24	75	-855	124
K	0	-13	3	0	0	37	-2
Ars	-5	529	616	55	119	581	187

K	0	
Ars	-5	1518

Covariance matrix for Group B

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V
Mo	3											
Cr	13	145										
P	1219	9782	750758									
Zn	123	884	70704	10610								

Pb	297	1922	161719	23547	53047		
Co	-3	16	23	50	-17	28	
Ni	2	41	2292	357	705	16	25
Fe	1	16	1061	37	88	0	2
Mg	-0	-0	-43	2	1	1	0
Cd	0	3	279	39	89	-0	1
Ti	-288	-3316	-230290	-1217	-5755	92	13
V	16	179	11815	978	2121	18	49
Be	0	1	56	13	25	1	1
Ca	0	-1	-47	7	17	-0	-0
Nb	-1	10	53	54	53	9	11
Cu	18	148	10705	1598	3439	28	75
Na	-1	-7	-514	-22	-54	0	-1
Zr	2	38	1717	472	913	17	34
Dy	-1	-5	-415	-31	-74	1	-1
Sc	-1	-4	-479	-20	-69	4	1
Y	-3	-33	-2264	-96	-227	-0	-5
Al	-0	2	-16	-18	-51	2	1
Mn	-188	1482	30411	-4383	-16480	1654	678
Sr	-25	-373	-22761	-720	-1530	-35	-64
La	-3	-17	-1507	-39	-130	4	3
Ce	-14	-39	-5571	-329	-1029	45	22
Ba	6	-371	-13776	4911	10847	-5	79
Li	3	98	5068	147	221	20	24
K	-0	-2	-205	-7	-21	1	0
Ars	7	53	4290	458	1048	0	12

Fe	3						
Mg	-0	0					
Cd	0	-0	0				
Ti	-673	42	-26	180471			
V	21	-0	4	-4454	224		
Be	-0	0	0	22	1	0	
Ca	-0	0	0	78	-2	0	0
Nb	0	0	-0	212	13	0	-0
Cu	5	1	5	-48	169	3	1
Na	-1	0	-0	297	-10	0	0
Zr	-1	1	1	1114	44	1	0
Dy	-1	0	-0	168	-7	-0	0
Sc	-1	0	-0	242	-5	0	0
Y	-5	0	-0	1316	-43	0	1
Al	0	0	-0	-102	3	0	-0
Mn	231	14	-23	-58895	1915	29	-56
Sr	-61	2	-3	14807	-495	-1	8
La	-4	0	-0	1019	-22	0	0
Ce	-9	2	-2	2579	-47	1	-0
Ba	-130	7	17	36885	-624	8	23
Li	15	-0	1	-3492	132	1	-2
K	-0	0	-0	124	-3	0	0
Ars	5	-0	2	-1084	62	0	-0

Nb	7						
Cu	21	290					
Na	0	-4	1				
Zr	18	101	0	59			
Dy	0	-4	0	-0	0		
Sc	2	-1	0	3	0	1	
Y	-0	-16	2	1	1	2	11
Al	1	-1	-0	0	0	0	-1
Mn	427	380	-82	186	1	194	-421
Sr	-21	-157	27	-10	15	14	121
La	4	2	2	11	1	2	7
Ce	24	0	5	46	4	11	18

Ba	-9	611	54	277	19	33	247
Li	13	42	-6	18	-3	-1	-30
K	0	-0	0	1	0	0	1
Ars	-1	62	-3	8	-2	-2	-11

Al	0						
Mn	179	129188					
Sr	-12	-6767	1444				
La	-0	-126	70	7			
Ce	3	2008	118	26	139		
Ba	-38	-16564	3176	155	155	11258	
Li	4	2327	-384	-13	2	-853	114
K	0	13	9	1	3	21	-2
Ars	-0	104	-103	-9	-36	35	21

K	0	
Ars	-1	27

Covariance matrix for Group P

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V
Mo	4											
Cr	42	1162										
P	2075	25530	2011690									
Zn	612	8474	315473	138468								
Pb	1426	37414	675856	366511	1861088							
Co	6	80	3712	931	2235	18						
Ni	23	423	13068	4263	14449	46	199					
Fe	2	36	1573	387	906	5	16					
Mg	-0	0	-44	-4	3	0	0					
Cd	1	12	541	303	763	2	7					
Ti	-361	3232	-340825	-75592	20780	-219	-358					
V	35	969	12785	6853	37416	68	375					
Be	1	7	413	109	242	2	6					
Ca	0	3	326	83	172	1	2					
Nb	1	13	140	60	346	4	8					
Cu	103	1346	75735	18789	38721	193	690					
Na	0	3	-88	33	68	-0	1					
Zr	-12	-210	-10595	-2776	-6325	-15	-94					
Dy	0	-1	-85	-13	14	1	1					
Sc	-0	-8	-929	-186	-412	1	-3					
Y	1	-5	-149	8	103	5	9					
Al	-1	-7	-692	-129	-244	-1	-3					
Mn	115	2340	116512	26123	79236	646	1280					
Sr	33	302	11779	5160	10834	40	233					
La	2	-0	-492	260	561	8	14					
Ce	4	2	-919	226	506	17	26					
Ba	315	5101	227173	94117	275653	535	2739					
Li	8	24	2106	724	2115	24	46					
K	-0	-2	-254	-28	-58	-0	-1					
Ars	36	261	25642	4540	6782	62	182					

Fe	2						
Mg	-0	0					
Cd	1	-0	1				
Ti	-98	40	-212	528230			
V	26	1	11	9897	1213		
Be	1	-0	0	-45	7	0	
Ca	0	-0	0	-114	0	0	0
Nb	1	-0	-0	602	22	1	-0
Cu	79	-1	42	-13723	739	25	12
Na	0	0	0	102	2	-0	0

Zr	-6	0	-7	3088	-79	-1	-4
Dy	0	0	-0	48	0	0	-0
Sc	-0	0	-0	850	15	0	-0
Y	1	0	-0	169	4	1	-0
Al	-0	0	-0	366	3	-0	-0
Mn	147	3	39	7675	1661	62	10
Sr	11	1	11	4518	332	9	9
La	1	0	0	-12	28	1	-0
Ce	2	0	-0	774	46	3	-1
Ba	181	-0	244	-48944	4131	54	98
Li	5	0	1	280	86	4	-0
K	-0	0	-0	76	0	-0	-0
Ars	24	-1	7	-6111	97	9	4

Nb	5						
Cu	25	6597					
Na	-0	3	0				
Zr	15	-489	-1	196			
Dy	1	7	-0	4	0		
Sc	2	-33	0	13	0	3	
Y	5	45	0	17	2	2	10
Al	1	-31	0	8	0	1	1
Mn	250	5126	4	-107	71	38	355
Sr	-3	574	7	-194	2	7	23
La	5	70	-0	18	2	3	9
Ce	15	99	-0	51	4	7	21
Ba	-109	12327	35	-2300	-7	-212	38
Li	15	157	-1	53	4	7	22
K	0	-7	0	1	0	0	-0
Ars	14	1037	-2	-117	2	-9	15

Al	1						
Mn	2	51899					
Sr	-2	-331	1320				
La	1	324	13	15			
Ce	3	1036	-6	28	67		
Ba	-124	20306	6019	24	-191	150137	
Li	2	968	33	29	63	135	83
K	0	-10	1	0	0	-26	-0
Ars	-9	1536	227	15	36	1945	83

K	0	
Ars	-4	471

Covariance matrix for Group R

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V
Mo	1											
Cr	14	3151										
P	63	-3797	463025									
Zn	94	6275	26796	45645								
Pb	114	801	48930	26054	68165							
Co	0	-14	393	68	143	11						
Ni	4	118	233	517	575	7	42					
Fe	0	2	136	24	43	1	1					
Mg	0	0	-13	3	-0	0	0					
Cd	0	2	70	70	57	0	1					
Ti	-129	-10381	28757	-32984	-19068	403	-1066					
V	3	-21	5054	476	1167	22	38					
Be	0	-2	48	7	19	1	1					
Ca	0	0	-26	28	5	-0	0					
Nb	-0	-39	390	-101	-34	2	-3					

Cu	49	1375	-8524	4608	4012	-16	274
Na	0	5	-89	35	16	-0	0
Zr	3	23	-1578	-582	-525	4	5
Dy	0	-5	-5	1	-6	1	0
Sc	-0	-14	172	-38	-33	3	1
Y	1	-19	-35	58	-8	4	3
Al	-0	-3	-34	-9	-16	0	-0
Mn	9	-958	20673	-2124	-4294	438	-15
Sr	-2	145	-9020	597	341	-4	61
La	1	-25	170	56	-2	6	4
Ce	1	-54	236	148	-176	14	5
Ba	73	1235	1995	14411	24933	39	419
Li	-0	-105	1773	-244	-28	11	3
K	0	3	-53	14	10	0	1
Ars	-2	-49	1353	-45	452	14	24

Fe	0						
Mg	0	0					
Cd	0	0	0				
Ti	120	-5	2	360167			
V	5	0	2	1465	238		
Be	0	0	0	56	2	0	
Ca	-0	0	0	-144	-3	-0	0
Nb	1	-0	0	1181	7	0	-1
Cu	13	1	10	-10213	116	2	1
Na	0	0	0	-3	-1	0	0
Zr	5	0	-1	5068	22	1	-3
Dy	0	0	0	236	1	0	-0
Sc	0	0	0	550	9	0	-0
Y	1	0	0	1130	5	1	-0
Al	0	0	0	219	2	0	-0
Mn	34	1	7	41921	941	28	-35
Sr	-6	2	1	-5486	-167	-1	8
La	1	0	0	1209	13	1	-1
Ce	2	0	1	2994	23	2	-1
Ba	19	2	33	-14056	353	9	8
Li	2	0	0	2611	46	1	-2
K	0	0	0	-22	-0	0	0
Ars	1	0	0	-249	33	1	0

Nb	7						
Cu	-43	5191					
Na	-0	9	0				
Zr	23	251	0	374			
Dy	1	0	0	4	0		
Sc	2	-14	-0	9	1	2	
Y	6	18	0	26	2	3	11
Al	1	1	0	5	0	1	1
Mn	245	-1739	-14	838	61	140	309
Sr	-37	392	5	-93	-2	-9	-6
La	6	1	0	24	2	3	11
Ce	16	-17	1	64	5	7	27
Ba	-73	4733	23	-68	7	-18	63
Li	13	-109	-1	44	3	7	12
K	-0	9	0	1	0	0	0
Ars	2	-149	-1	-1	-1	2	-4

Al	0					
Mn	35	40265				
Sr	-1	-2447	918			
La	1	410	-11	13		
Ce	3	975	-39	30	80	
Ba	0	-3033	1328	47	19	15674

Li	2	659	-95	14	35	-170	43
K	0	-1	3	0	1	17	-0
Ars	-0	-97	130	-0	-2	60	15

K	0	
Ars	-0	120

Appendix 2

Covariance Matrices for Expanded Pollution Sources

Pooled Covariance Matrix

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V	Be
Mo	8												
Cr	20	1047											
P	521	5201	819602										
Zn	155	3246	87259	48214									
Pb	346	5475	190210	69967	275386								
Co	7	32	1186	310	661	22							
Ni	28	138	2824	920	2418	36	141						
Fe	4	13	536	89	243	5	17						
Mg	-0	0	-16	-1	-3	0	0						
Cd	0	3	164	78	138	1	1						
Ti	-276	-1072	49166	-17019	2911	738	-204						
V	16	186	7737	1341	5247	44	122						
Be	1	2	101	30	69	2	4						
Ca	0	1	45	23	24	-0	0						
Nb	0	-6	385	-17	84	3	3						
Cu	297	932	10063	5306	9610	259	1218						
Na	-0	1	-144	17	10	-0	-1						
Zr	-5	6	-801	-403	-806	18	-3						
Dy	0	-1	-121	-8	-13	2	2						
Sc	0	2	127	-19	-12	4	4						
Y	2	-1	-545	9	-8	10	13						
Al	-0	-1	-78	-11	-19	0	-1						
Mn	76	1123	25324	7769	11533	1142	663						
Sr	36	74	-6058	774	1647	13	149						
La	3	0	-913	40	38	12	16						
Ce	5	0	-1360	61	20	27	32						
Ba	109	944	9535	17337	44457	36	473						
Li	10	14	2465	162	589	29	51						
K	-0	-0	-152	-5	-20	-0	-1						
Ars	77	163	7050	1198	3089	77	285						
Fe	3												
Mg	-0	0											
Cd	0	-0	0										
Ti	131	36	-37	494432									
V	16	1	2	6083	415								
Be	1	0	0	5	3	0							
Ca	-0	0	0	-80	-2	-0	0						
Nb	1	0	-0	1275	18	0	-0						
Cu	160	-2	7	-12413	539	32	4						
Na	-0	0	0	-35	-3	-0	0						
Zr	2	1	-1	6070	75	1	-2						
Dy	0	0	-0	221	2	0	-0						
Sc	1	0	-0	940	18	0	-0						
Y	2	0	0	995	12	1	-0						
Al	-0	0	-0	156	2	0	-0						
Mn	119	14	27	42049	1276	60	-12						
Sr	11	-0	2	-5943	-132	4	5						
La	1	0	0	330	10	2	-0						
Ce	4	1	0	2343	36	3	-1						
Ba	17	-3	42	-46168	-219	14	19						
Li	9	0	-0	3636	104	3	-1						
K	-0	0	-0	-63	-2	-0	-0						
Ars	44	-1	1	-1967	168	10	1						
Nb	7												
Cu	-11	14970											

Na	-0	-7	0				
Zr	23	-172	-1	268			
Dy	1	11	0	5	1		
Sc	3	0	-0	16	1	3	
Y	5	93	0	26	3	4	17
Al	1	-12	0	4	0	0	0
Mn	189	2164	-18	1152	120	242	700
Sr	-29	1675	6	-172	-1	-13	8
La	3	120	0	22	4	4	20
Ce	14	205	0	70	7	11	37
Ba	-150	4950	32	-818	-24	-121	-67
Li	16	313	-2	78	4	13	21
K	-0	-8	0	-0	0	-0	0
Ars	8	2929	-3	-41	3	5	25
Al	1						
Mn	-5	174467					
Sr	-0	-2083	1016				
La	0	864	14	30			
Ce	1	1843	2	47	104		
Ba	-6	-4742	2662	10	-174	43846	
Li	2	1263	-69	20	56	-564	88
K	0	-7	3	0	0	20	-1
Ars	-3	399	417	31	61	718	120
K	0						
Ars	-4	972					

Covariance matrix for Group AD

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V	Be
Mo	1												
Cr	5	108											
P	368	899	579780										
Zn	47	424	31265	39679									
Pb	169	1291	76825	26876	87203								
Co	2	10	740	208	436	14							
Ni	6	42	1291	419	1397	14	47						
Fe	1	4	335	15	167	2	5						
Mg	-0	0	-15	1	3	0	0						
Cd	-0	0	118	50	60	1	1						
Ti	135	-596	49032	-16320	-7671	1060	788						
V	9	55	5248	169	2040	25	80						
Be	0	1	74	16	36	1	2						
Ca	-0	0	61	24	21	0	0						
Nb	1	-1	182	-50	24	5	6						
Cu	20	272	6114	3610	5689	52	165						
Na	-0	-0	-138	27	4	-0	-0						
Zr	2	4	504	-214	-463	18	16						
Dy	0	-0	-46	9	33	2	2						
Sc	0	1	158	-18	27	4	4						
Y	1	-1	-228	70	185	9	9						
Al	-0	1	-45	-1	-50	1	1						
Mn	-8	-62	23530	4193	7733	919	379						
Sr	-1	13	-1565	1196	1566	3	16						
La	0	3	-727	75	199	10	10						
Ce	2	6	-799	212	582	28	29						
Ba	4	437	-11027	10446	18771	1	166						
Li	5	10	2560	40	540	25	30						
K	-0	0	-148	5	-20	-0	-0						
Ars	16	88	4341	209	1777	26	55						
Fe	1												
Mg	0	0											
Cd	-0	0	0										

Ti	308	35	-69	420328			
V	10	1	-1	5584	379		
Be	0	0	-0	86	2	0	
Ca	-0	0	0	-66	-2	-0	0
Nb	2	0	-0	1469	20	1	-0
Cu	10	0	7	88	242	5	5
Na	-0	0	0	-51	-2	-0	0
Zr	5	1	-2	6661	92	2	-1
Dy	0	0	0	284	5	0	-0
Sc	1	0	-0	839	17	0	-0
Y	1	0	0	1345	21	1	-0
Al	0	0	-0	133	2	0	-0
Mn	113	13	50	62428	516	44	3
Sr	-5	-0	5	-5085	-124	0	5
La	1	0	0	1160	25	1	-0
Ce	4	1	0	3568	67	3	-0
Ba	-18	-1	24	-45781	-630	1	17
Li	7	1	-0	4822	95	3	-1
K	-0	0	-0	-88	-2	-0	0
Ars	11	-0	-1	1978	133	5	-1
Nb	8						
Cu	-5	2190					
Na	-0	2	0				
Zr	27	-57	-1	164			
Dy	1	1	0	5	1		
Sc	3	-4	-0	15	1	3	
Y	7	14	0	26	3	5	16
Al	0	-1	0	3	-0	1	-0
Mn	265	560	-5	1198	134	246	695
Sr	-20	354	4	-127	0	-10	5
La	7	27	0	26	4	5	18
Ce	20	34	0	73	8	12	40
Ba	-157	2110	23	-740	-17	-86	-57
Li	22	8	-1	91	5	14	25
K	-0	-0	0	-1	-0	-0	-0
Ars	19	324	-2	33	3	6	17
Al	1						
Mn	16	101129					
Sr	1	-166	418				
La	-0	765	15	25			
Ce	-0	2038	-1	50	121		
Ba	-5	-2524	1531	23	-143	16285	
Li	3	1274	-58	24	71	-527	96
K	0	-6	2	0	0	21	-1
Ars	-0	290	-31	16	49	-72	71
K	0						
Ars	-2	331					

Covariance matrix for Group AS

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V	Be
Mo	20												
Cr	23	162											
P	-167	4253	642030										
Zn	26	676	38396	17093									
Pb	51	877	56863	14767	41193								
Co	15	57	477	202	214	36							
Ni	72	127	900	232	357	75	297						
Fe	11	19	303	34	77	10	41						
Mg	-0	1	20	3	1	0	-0						
Cd	-0	1	31	15	28	0	-1						
Ti	-752	4910	284886	15224	21242	1477	-353						
V	24	207	9515	971	988	72	155						

Be	2	3	-54	12	19	3	8
Ca	0	-0	-0	5	2	0	0
Nb	0	10	863	63	170	4	4
Cu	863	1049	-6008	1365	3832	701	3336
Na	-1	-3	-224	-11	-17	-1	-4
Zr	-15	71	1708	67	5	44	-6
Dy	1	4	-100	8	8	4	5
Sc	0	18	637	58	56	8	8
Y	7	23	-652	60	36	20	33
Al	-1	-1	-122	-16	-23	-0	-3
Mn	230	3840	2075	19763	11046	2082	1624
Sr	104	-40	-15564	-1174	-1082	21	317
La	9	21	-1691	62	-26	23	39
Ce	15	51	-1720	202	197	47	75
Ba	128	-444	-76435	-1470	7395	-202	73
Li	23	99	3419	443	575	56	118
K	-1	-2	-195	-12	-18	-1	-3
Ars	210	250	-468	66	524	162	723
Fe	6						
Mg	-0	0					
Cd	-0	0	0				
Ti	175	75	12	759516			
V	26	1	0	11406	417		
Be	1	-0	-0	-96	3	0	
Ca	0	0	0	-34	-1	0	0
Nb	2	0	0	1627	28	0	-0
Cu	466	-6	-10	-29250	1122	88	6
Na	-1	-0	-0	-140	-7	-0	0
Zr	-0	2	-0	8966	152	-0	-1
Dy	1	0	0	179	4	0	0
Sc	2	0	0	1583	34	0	-0
Y	4	0	0	679	20	2	0
Al	-0	0	-0	172	-1	-0	-0
Mn	219	32	47	51978	2527	120	-1
Sr	39	-2	-3	-13942	-295	9	4
La	3	0	0	-1376	-2	3	0
Ce	9	1	0	999	44	5	0
Ba	-22	-8	2	-89514	-1931	16	10
Li	20	1	0	5736	181	3	-0
K	-1	0	-0	-167	-6	-0	0
Ars	115	-1	-3	-5142	296	21	1
Nb	9						
Cu	-1	40688					
Na	-1	-33	0				
Zr	26	-570	-3	369			
Dy	1	41	-0	6	1		
Sc	3	20	-0	26	1	5	
Y	3	290	-0	30	5	6	27
Al	0	-36	0	4	0	0	0
Mn	127	6932	-37	1972	220	437	1418
Sr	-49	4647	9	-356	-2	-36	6
La	-2	367	-0	16	7	4	34
Ce	7	672	-1	78	10	13	53
Ba	-221	5820	55	-1328	-35	-231	-109
Li	19	1053	-4	117	6	21	31
K	-0	-31	0	-1	-0	-0	-0
Ars	4	8765	-8	-123	10	10	73
Al	1						
Mn	-27	398213					
Sr	-2	-3493	1600				
La	-0	1819	25	57			
Ce	1	3104	21	74	132		
Ba	7	-12879	4535	135	-99	30319	

Li	0	2254	-111	23	75	-1115	139
K	0	-18	5	0	0	48	-3
Ars	-8	765	1089	83	169	1045	269
K	0						
Ars	-7	2381					

Covariance matrix for Group BS

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V
Mo	3											
Cr	13	145										
P	1219	9782	750758									
Zn	123	884	70704	10610								
Pb	297	1922	161719	23547	53047							
Co	-3	16	23	50	-17	28						
Ni	2	41	2292	357	705	16	25					
Fe	1	16	1061	37	88	0	2					
Mg	-0	-0	-43	2	1	1	0					
Cd	0	3	279	39	89	-0	1					
Ti	-288	-3316	-230290	-1217	-5755	92	13					
V	16	179	11815	978	2121	18	49					
Be	0	1	56	13	25	1	1					
Ca	0	-1	-47	7	17	-0	-0					
Nb	-1	10	53	54	53	9	11					
Cu	18	148	10705	1598	3439	28	75					
Na	-1	-7	-514	-22	-54	0	-1					
Zr	2	38	1717	472	913	17	34					
Dy	-1	-5	-415	-31	-74	1	-1					
Sc	-1	-4	-479	-20	-69	4	1					
Y	-3	-33	-2264	-96	-227	-0	-5					
Al	-0	2	-16	-18	-51	2	1					
Mn	-188	1482	30411	-4383	-16480	1654	678					
Sr	-25	-373	-22761	-720	-1530	-35	-64					
La	-3	-17	-1507	-39	-130	4	3					
Ce	-14	-39	-5571	-329	-1029	45	22					
Ba	6	-371	-13776	4911	10847	-5	79					
Li	3	98	5068	147	221	20	24					
K	-0	-2	-205	-7	-21	1	0					
Ars	7	53	4290	458	1048	0	12					
Fe	3											
Mg	-0	0										
Cd	0	-0	0									
Ti	-673	42	-26	180471								
V	21	-0	4	-4454	224							
Be	-0	0	0	22	1	0						
Ca	-0	0	0	78	-2	0	0					
Nb	0	0	-0	212	13	0	-0					
Cu	5	1	5	-48	169	3	1					
Na	-1	0	-0	297	-10	0	0					
Zr	-1	1	1	1114	44	1	0					
Dy	-1	0	-0	168	-7	-0	0					
Sc	-1	0	-0	242	-5	0	0					
Y	-5	0	-0	1316	-43	0	1					
Al	0	0	-0	-102	3	0	-0					
Mn	231	14	-23	-58895	1915	29	-56					
Sr	-61	2	-3	14807	-495	-1	8					
La	-4	0	-0	1019	-22	0	0					
Ce	-9	2	-2	2579	-47	1	-0					
Ba	-130	7	17	36885	-624	8	23					
Li	15	-0	1	-3492	132	1	-2					
K	-0	0	-0	124	-3	0	0					
Ars	5	-0	2	-1084	62	0	-0					
Nb	7											

Cu	21	290					
Na	0	-4	1				
Zr	18	101	0	59			
Dy	0	-4	0	-0	0		
Sc	2	-1	0	3	0	1	
Y	-0	-16	2	1	1	2	11
Al	1	-1	-0	0	0	0	-1
Mn	427	380	-82	186	1	194	-421
Sr	-21	-157	27	-10	15	14	121
La	4	2	2	11	1	2	7
Ce	24	0	5	46	4	11	18
Ba	-9	611	54	277	19	33	247
Li	13	42	-6	18	-3	-1	-30
K	0	-0	0	1	0	0	1
Ars	-1	62	-3	8	-2	-2	-11
Al	0						
Mn	179	129188					
Sr	-12	-6767	1444				
La	-0	-126	70	7			
Ce	3	2008	118	26	139		
Ba	-38	-16564	3176	155	155	11258	
Li	4	2327	-384	-13	2	-853	114
K	0	13	9	1	3	21	-2
Ars	-0	104	-103	-9	-36	35	21
K	0						
Ars	-1	27					

Covariance matrix for Group CD

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V
Mo	6											
Cr	48	444										
P	2914	23981	1955877									
Zn	679	6183	405341	123080								
Pb	1640	14417	994184	252758	630632							
Co	10	88	5122	1269	3194	32						
Ni	27	233	12658	3334	7536	51	145					
Fe	3	27	1807	389	1022	6	16					
Mg	-0	-1	-89	-19	-39	-0	-1					
Cd	1	12	838	233	481	2	6					
Ti	-349	-2986	-338566	-78070	-136021	-761	-1764					
V	24	210	13499	3331	7764	51	142					
Be	1	10	593	147	374	3	6					
Ca	0	3	208	53	93	0	2					
Nb	0	3	-409	-68	30	-1	3					
Cu	121	1105	70223	17603	42366	247	580					
Na	0	1	-44	23	39	0	0					
Zr	-5	-37	-5701	-1224	-2910	-9	-20					
Dy	-0	-3	-510	-75	-123	-1	-2					
Sc	-0	0	-584	-105	-45	1	0					
Y	-1	-2	-1748	-207	-203	1	-2					
Al	0	3	247	27	111	1	2					
Mn	31	214	45146	-1148	24222	1021	-109					
Sr	35	283	12591	4108	9461	62	178					
La	1	11	-1156	-67	146	4	7					
Ce	-1	-5	-3844	-596	-591	6	-4					
Ba	352	3162	213234	54185	116814	609	1465					
Li	9	68	2836	687	2321	18	46					
K	-0	-3	-219	-43	-103	-0	-2					
Ars	73	602	37069	8475	21105	137	342					
Fe	2											
Mg	-0	0										

Cd	1	-0	1				
Ti	-140	18	-184	302431			
V	18	-1	6	-755	252		
Be	1	-0	0	-33	7	0	
Ca	0	-0	0	-132	0	0	0
Nb	1	0	-0	814	7	0	-0
Cu	74	-3	36	-8534	596	27	7
Na	0	0	0	40	0	0	-0
Zr	-2	0	-3	4083	19	-1	-3
Dy	-0	0	-0	291	-1	0	-0
Sc	0	0	-0	546	5	0	-0
Y	0	0	-1	1256	5	1	-1
Al	0	-0	0	-63	5	0	-0
Mn	23	7	-9	5455	-202	10	-23
Sr	21	-0	6	-1778	130	10	2
La	1	0	-1	1159	8	1	-1
Ce	0	0	-2	2877	-4	1	-2
Ba	153	-9	112	-58571	1256	51	28
Li	7	-0	1	1059	75	3	-0
K	-0	0	-0	47	-1	-0	-0
Ars	46	-2	16	-4143	356	18	4
Nb	4						
Cu	1	3318					
Na	0	2	0				
Zr	14	-178	1	137			
Dy	1	-12	0	5	1		
Sc	2	-5	0	11	1	2	
Y	4	-29	1	24	3	6	16
Al	0	8	-0	1	-0	0	-1
Mn	-27	1109	-5	572	20	89	142
Sr	3	626	2	-27	3	10	25
La	4	8	1	21	3	6	15
Ce	9	-51	1	52	6	12	27
Ba	-170	9013	-1	-1212	-78	-122	-314
Li	7	152	0	28	3	7	16
K	0	-7	0	1	0	0	0
Ars	5	1631	-0	-72	-6	2	-7
Al	1						
Mn	-137	130163					
Sr	2	-1343	489				
La	-1	137	47	17			
Ce	-0	651	60	31	70		
Ba	6	-3833	1026	-307	-697	137922	
Li	2	29	84	17	28	-142	45
K	0	8	-1	0	1	-24	-0
Ars	5	190	465	19	-9	3464	131
K	0						
Ars	-4	1033					

Covariance matrix for Group PD

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V
Mo	4											
Cr	39	1242										
P	1792	22741	2040470									
Zn	476	7942	235686	116362								
Pb	1066	40838	453474	323259	2033867							
Co	5	66	3197	606	1538	13						
Ni	19	407	10352	3348	13336	36	171					
Fe	2	37	1539	346	800	4	15					
Mg	-0	-0	-63	-5	5	-0	-0					
Cd	1	8	272	244	627	1	4					

Ti	-1	3750	-263154	-12502	224056	324	1229
V	36	1039	8056	6905	43612	54	383
Be	1	6	320	57	108	2	4
Ca	0	1	276	59	104	0	1
Nb	1	14	114	9	247	4	9
Cu	68	1093	62188	11695	19219	128	437
Na	0	2	-107	49	98	0	1
Zr	-5	-118	-7866	-1708	-3535	1	-35
Dy	-0	-3	-225	-75	-119	1	-0
Sc	0	-12	-1003	-134	-242	1	-2
Y	-0	-14	-786	-272	-539	5	3
Al	-0	-5	-627	-75	-81	-0	-1
Mn	66	1889	106427	10952	55846	404	920
Sr	30	163	8406	4900	9172	58	189
La	1	-12	-1533	-124	-344	5	5
Ce	1	-18	-2387	-459	-1160	12	13
Ba	171	4160	165758	77508	249814	303	1883
Li	5	2	303	-78	345	18	30
K	-0	-2	-249	-12	-17	-0	-1
Ars	31	193	25220	2948	1378	54	131
Fe	2						
Mg	-0	0					
Cd	0	-0	1				
Ti	45	19	-77	319816			
V	25	0	9	11178	1328		
Be	0	-0	-0	63	7	0	
Ca	0	-0	0	-100	-1	0	0
Nb	2	0	-1	843	27	1	-0
Cu	70	-1	24	-616	529	15	6
Na	0	0	0	37	3	0	-0
Zr	-2	1	-5	4144	4	1	-2
Dy	-0	0	-0	164	1	0	-0
Sc	-0	0	-0	639	13	0	-0
Y	0	0	-1	749	5	1	-0
Al	-0	0	-0	300	6	0	-0
Mn	134	-0	5	23197	1238	47	3
Sr	13	-0	10	1768	350	9	6
La	-0	0	-1	1086	32	1	-0
Ce	1	0	-2	2484	55	2	-1
Ba	127	-4	208	-10385	3825	12	79
Li	4	0	-2	2748	91	3	-0
K	-0	0	0	54	1	-0	-0
Ars	24	-1	2	-3913	43	7	3
Nb	6						
Cu	18	5918					
Na	0	5	0				
Zr	20	-234	0	145			
Dy	1	-2	0	6	0		
Sc	2	-20	0	16	1	3	
Y	6	4	0	28	2	3	11
Al	1	-21	0	7	0	1	1
Mn	246	2742	11	622	65	20	345
Sr	11	324	2	-39	2	11	26
La	5	0	0	29	2	5	9
Ce	15	-17	0	77	4	10	22
Ba	-134	6806	34	-1248	-40	-198	-120
Li	17	2	0	79	4	12	23
K	0	-5	0	1	0	0	0
Ars	12	730	-2	-65	-0	-5	6
Al	1						
Mn	12	40039					
Sr	1	222	873				
La	2	138	41	13			

Ce	4	768	42	24	62		
Ba	-94	13562	4685	-206	-575	157005	
Li	4	781	102	25	59	-329	79
K	0	-7	-1	0	1	-22	0
Ars	-7	1144	222	-4	3	542	57

K	0	
Ars	-4	453

Covariance matrix for Group PS

	Mo	Cr	P	Zn	Pb	Co	Ni
Mo	0						
Cr	-1	433					
P	-35	6314	233977				
Zn	53	349	14274	23313			
Pb	163	1559	39811	48814	136852		
Co	-0	47	444	386	519	26	
Ni	1	133	1690	407	1282	20	48
Fe	-0	7	246	34	64	2	2
Mg	-0	3	21	-1	-8	0	1
Cd	0	2	45	29	73	1	1
Ti	-87	18814	279047	-7986	20757	512	5304
V	-5	447	10524	-60	-244	64	122
Be	0	0	-23	39	98	1	1
Ca	-0	1	-47	-16	-44	-0	0
Nb	1	7	142	232	698	5	7
Cu	9	145	2064	2978	7990	67	97
Na	-0	5	-16	-22	-31	-1	1
Zr	-0	-230	-1284	-102	-502	-17	-75
Dy	0	-2	-42	60	139	0	0
Sc	-1	30	492	3	-107	6	8
Y	1	-15	-282	230	511	0	-1
Al	-0	4	4	-6	-56	1	1
Mn	49	1818	19685	38418	57605	1255	1023
Sr	-5	363	-4281	-3556	-6552	-119	65
La	1	-25	-534	451	891	13	0
Ce	3	-15	-772	1124	2680	27	17
Ba	6	647	-12410	47	-3980	-3	221
Li	1	-49	-83	772	1553	22	-4
K	-0	1	-75	-14	-30	-0	0
Ars	5	35	319	1127	3876	13	35

	Fe	Mg	Cd	Ti	V	Be	
Fe	0						
Mg	0	0					
Cd	0	0	0				
Ti	167	133	36	961963			
V	14	2	2	17941	682		
Be	0	-0	0	-50	-2	0	
Ca	-0	0	-0	125	-1	-0	0
Nb	1	-0	0	-198	3	1	-0
Cu	7	-0	5	992	65	6	-3
Na	-0	0	-0	372	1	-0	0
Zr	-0	-2	-0	-11494	-166	-0	-2
Dy	-0	-0	0	-158	-6	0	-0
Sc	1	0	0	1203	40	-0	0
Y	-0	-0	0	-807	-32	0	-0
Al	0	0	0	159	4	-0	0
Mn	91	14	48	24253	1801	63	-12
Sr	-21	6	-6	31846	-53	-6	14
La	1	-0	1	-2367	-30	1	-1
Ce	2	-0	2	-3183	-48	3	-1
Ba	-27	12	-5	47188	-58	2	25
Li	2	-1	1	-4725	-30	2	-2

K	-0	0	-0	57	-1	-0	0
Ars	1	-1	2	-460	-32	3	-2
Nb	4						
Cu	46	533					
Na	-0	-3	0				
Zr	0	-45	-6	189			
Dy	1	8	-0	2	0		
Sc	-1	1	0	-14	-0	3	
Y	2	27	-0	8	1	-2	4
Al	-0	-1	0	-3	-0	1	-0
Mn	300	4504	-20	-1316	64	216	246
Sr	-61	-545	27	-497	-9	12	-30
La	6	69	-1	20	1	-2	5
Ce	17	188	-2	14	3	-3	12
Ba	-58	-317	45	-1132	-10	34	-19
Li	11	119	-4	65	3	-2	9
K	-0	-1	0	-2	-0	0	-0
Ars	22	239	-1	-0	4	-5	14
Al	0						
Mn	49	95717					
Sr	5	-5068	2833				
La	-0	799	-163	17			
Ce	-1	1844	-288	35	83		
Ba	14	9668	4107	-129	-145	10723	
Li	-0	1116	-388	30	58	-535	65
K	0	-5	10	-0	-0	20	-1
Ars	-2	908	-210	28	83	-315	50
K	0						
Ars	-1	131					

Covariance matrix for Group RD

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V
Mo	1											
Cr	7	3410										
P	162	-190	475966									
Zn	106	7130	32637	53824								
Pb	133	811	53444	29782	78961							
Co	1	-6	153	60	126	11						
Ni	3	97	457	516	575	7	38					
Fe	0	-1	162	27	51	1	1					
Mg	0	0	-17	3	-1	0	0					
Cd	0	3	66	80	67	0	1					
Ti	-115	-10496	-2165	-37321	-19225	427	-1117					
V	5	24	5260	560	1444	21	46					
Be	0	-2	41	7	18	1	1					
Ca	0	0	-41	31	5	-0	0					
Nb	-0	-38	242	-101	-48	2	-3					
Cu	41	590	-720	4298	4638	9	239					
Na	0	4	-74	41	22	-0	0					
Zr	-0	-137	-316	-645	-354	12	1					
Dy	0	-7	-33	-3	-15	1	0					
Sc	-0	-13	96	-56	-50	3	0					
Y	1	-28	-148	50	-44	4	1					
Al	-0	-4	-41	-12	-16	0	-0					
Mn	9	-805	8725	-2808	-8194	469	-67					
Sr	-4	49	-9776	438	411	-3	61					
La	1	-31	-27	32	-57	6	3					
Ce	1	-67	-160	113	-308	14	3					
Ba	75	697	8559	15895	29356	48	404					
Li	0	-98	1514	-313	-116	10	2					
K	0	1	-39	15	13	0	1					

Ars	-3	-38	927	-99	463	13	26
Fe	0						
Mg	0	0					
Cd	0	0	0				
Ti	133	-5	0	397155			
V	6	-0	2	1516	270		
Be	0	0	0	65	2	0	
Ca	-0	0	0	-175	-4	-0	0
Nb	1	-0	0	1263	8	0	-1
Cu	8	1	12	-8362	268	2	1
Na	0	0	0	9	-1	0	0
Zr	4	0	-0	5825	51	2	-4
Dy	0	0	0	276	1	0	-0
Sc	0	0	0	613	9	0	-0
Y	1	0	0	1321	6	1	-0
Al	0	0	0	257	2	0	-0
Mn	32	2	8	46805	1013	27	-43
Sr	-7	2	0	-6034	-200	-1	9
La	1	0	1	1438	13	1	-1
Ce	2	0	1	3552	22	2	-2
Ba	22	2	38	-11571	488	10	9
Li	3	0	0	2782	50	1	-2
K	0	0	0	-7	0	0	-0
Ars	1	0	-0	-568	31	1	0
Nb	7						
Cu	-38	3846					
Na	-0	7	0				
Zr	26	-53	-0	162			
Dy	1	-0	0	5	1		
Sc	2	-9	0	12	1	2	
Y	6	8	1	26	3	3	13
Al	1	-2	0	5	0	1	1
Mn	231	-1351	-9	915	66	154	325
Sr	-38	222	5	-121	-3	-11	-8
La	7	1	0	28	3	4	13
Ce	18	-24	1	75	6	8	31
Ba	-59	4183	25	-227	7	-19	63
Li	14	-68	-1	62	3	7	14
K	-0	6	0	0	0	0	0
Ars	0	-133	-1	11	-1	2	-6
Al	0						
Mn	43	43334					
Sr	-2	-2695	1066				
La	1	432	-14	15			
Ce	4	1089	-48	34	92		
Ba	1	-3339	1340	43	-2	17338	
Li	2	665	-109	16	39	-170	44
K	0	3	3	0	1	16	-0
Ars	-0	-276	167	-3	-8	82	11
K	0						
Ars	-0	138					

Covariance matrix for Group RS

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V	Be
Mo	1												
Cr	36	2134											
P	-207	-17228	352004										
Zn	71	3360	-10554	10576									
Pb	37	818	32109	11351	25208								
Co	0	-37	1118	53	226	8							
Ni	6	214	-641	573	616	8	62						

Fe	0	11	77	20	13	0	1
Mg	0	2	-10	4	1	0	0
Cd	0	5	4	14	17	0	1
Ti	-180	-10207	155300	-17911	-19678	279	-902
V	-0	-164	2723	-141	26	19	4
Be	0	-0	102	11	26	1	1
Ca	0	3	-13	9	7	0	1
Nb	-1	-54	1318	-68	24	2	-4
Cu	76	4670	-35613	7513	1543	-95	437
Na	0	9	-160	14	-8	-1	1
Zr	10	611	-3478	249	-1304	-11	20
Dy	0	0	120	18	31	1	1
Sc	-0	-11	365	13	42	2	2
Y	1	20	558	113	148	4	9
Al	0	5	-2	6	-14	-0	-0
Mn	1	-1886	80580	1685	12596	356	208
Sr	11	632	-8058	1058	49	-17	65
La	1	6	879	138	240	6	9
Ce	2	12	1653	255	389	11	15
Ba	73	3737	-28465	8571	7132	-6	515
Li	-1	-133	2797	9	352	14	8
K	0	10	-113	15	-1	-0	1
Ars	0	-78	2574	74	436	14	15
Fe	0						
Mg	0	0					
Cd	0	0	0				
Ti	77	-8	-2	219893			
V	0	0	-0	1101	76		
Be	0	0	0	25	1	0	
Ca	0	0	0	-24	-0	0	0
Nb	1	-0	0	932	7	0	-0
Cu	30	4	12	-17984	-385	-1	7
Na	-0	0	0	-56	-2	-0	0
Zr	6	1	0	2579	-27	-0	0
Dy	0	0	0	73	1	0	0
Sc	0	0	0	286	5	0	-0
Y	1	0	0	379	7	0	0
Al	0	0	0	66	0	0	0
Mn	45	-1	9	24030	812	36	-1
Sr	0	0	1	-3679	-67	-1	1
La	1	0	0	262	13	1	0
Ce	2	0	0	692	22	1	0
Ba	7	4	11	-26158	-250	4	8
Li	1	-0	0	2003	28	1	-0
K	0	0	0	-92	-1	-0	0
Ars	1	0	1	1038	29	1	0
Nb	8						
Cu	-88	10732					
Na	-1	21	0				
Zr	4	1259	1	1167			
Dy	0	2	-0	1	0		
Sc	1	-19	-0	3	0	1	
Y	3	50	-0	22	1	1	5
Al	0	16	0	4	0	0	0
Mn	304	-4110	-39	293	43	101	255
Sr	-31	1328	5	106	-0	-6	4
La	3	16	-0	17	1	2	5
Ce	7	47	-0	35	2	3	10
Ba	-136	7623	16	699	8	-20	68
Li	10	-271	-1	-18	2	5	9
K	-0	20	0	2	-0	-0	-0
Ars	11	-163	-1	-21	2	3	9
Al	0						

Mn	4	29029					
Sr	1	-1396	293				
La	0	353	-4	8			
Ce	1	562	-8	14	30		
Ba	-1	-1767	1327	63	107	9363	
Li	0	691	-44	10	19	-184	41
K	0	-17	3	-0	-0	19	-1
Ars	-0	739	-36	10	22	-50	32
K	0						
Ars	-1	42					

Appendix 3

Covariance Matrices for Downtown Pollution Sources

Pooled Covariance Matrix

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V	Be
Mo	2												
Cr	18	1450											
P	916	7255	965204										
Zn	231	4681	122056	68612									
Pb	524	8362	274570	104149	422853								
Co	3	25	1603	390	944	16							
Ni	10	140	4105	1324	3674	21	76						
Fe	1	10	681	125	354	3	7						
Mg	-0	-0	-34	-3	-6	0	0						
Cd	0	4	244	117	206	1	2						
Ti	-52	-3884	-74854	-33766	-4722	434	-256						
V	14	195	7031	1697	8047	32	117						
Be	0	2	183	41	97	1	2						
Ca	0	1	78	36	38	0	0						
Nb	1	-11	92	-63	28	3	3						
Cu	51	629	21875	7282	13363	80	296						
Na	-0	2	-96	34	28	-0	0						
Zr	-1	-67	-1903	-725	-1234	9	-2						
Dy	-0	-3	-144	-20	-30	1	0						
Sc	0	-6	-138	-61	-46	3	1						
Y	0	-12	-533	-28	-52	5	4						
Al	-0	-1	-66	-10	-14	1	0						
Mn	14	-26	32565	1700	11124	716	209						
Sr	8	91	-704	1903	3487	18	81						
La	1	-9	-661	11	33	7	7						
Ce	1	-24	-1304	-46	-147	17	12						
Ba	109	1466	56701	28313	68400	160	688						
Li	4	-18	1963	15	597	18	23						
K	-0	-0	-135	-2	-23	-0	-0						
Ars	21	146	11439	1875	4612	44	104						
Fe	1												
Mg	-0	0											
Cd	0	-0	0										
Ti	137	16	-66	379321									
V	12	0	3	3786	438								
Be	0	0	0	55	4	0							
Ca	-0	0	0	-119	-2	-0	0						
Nb	1	0	-0	1206	15	1	-0						
Cu	28	-0	16	-4410	348	9	4						
Na	-0	0	0	-4	-0	0	0						
Zr	3	0	-2	5611	54	1	-2						
Dy	0	0	-0	267	2	0	-0						
Sc	0	0	-0	686	12	0	-0						
Y	1	0	-0	1247	11	1	-0						
Al	0	0	-0	163	3	0	-0						
Mn	72	6	20	42257	654	32	-17						
Sr	1	1	4	-3981	-47	2	6						
La	1	0	0	1247	19	1	-0						
Ce	2	0	-0	3307	38	2	-1						
Ba	43	-2	67	-31722	642	14	24						
Li	5	0	-0	3206	75	2	-1						
K	-0	0	-0	-19	-1	-0	-0						
Ars	15	-0	3	-700	125	6	1						
Nb	6												
Cu	-12	3426											

Na	-0	4	0				
Zr	23	-99	-0	156			
Dy	1	-2	0	5	1		
Sc	2	-8	0	13	1	2	
Y	6	4	0	26	3	4	14
Al	1	-2	0	4	0	1	0
Mn	200	269	-4	920	82	159	428
Sr	-18	352	4	-97	-0	-4	7
La	6	11	0	26	3	5	15
Ce	17	-7	1	71	7	10	33
Ba	-122	4609	21	-708	-22	-83	-68
Li	16	6	-0	68	4	10	19
K	-0	0	0	0	0	0	0
Ars	9	443	-1	-5	-1	3	3
Al	1						
Mn	-2	78498					
Sr	0	-1194	711				
La	1	462	14	19			
Ce	2	1311	-1	38	95		
Ba	-12	-1010	1774	-56	-244	55347	
Li	3	786	-31	20	51	-312	67
K	0	-1	1	0	1	6	-1
Ars	-0	184	155	8	13	670	59
K	0						
Ars	-2	401					

Covariance matrix for Group A

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V	Be
Mo	1												
Cr	5	108											
P	368	899	579780										
Zn	47	424	31265	39679									
Pb	169	1291	76825	26876	87203								
Co	2	10	740	208	436	14							
Ni	6	42	1291	419	1397	14	47						
Fe	1	4	335	15	167	2	5						
Mg	-0	0	-15	1	3	0	0						
Cd	-0	0	118	50	60	1	1						
Ti	135	-596	49032	-16320	-7671	1060	788						
V	9	55	5248	169	2040	25	80						
Be	0	1	74	16	36	1	2						
Ca	-0	0	61	24	21	0	0						
Nb	1	-1	182	-50	24	5	6						
Cu	20	272	6114	3610	5689	52	165						
Na	-0	-0	-138	27	4	-0	-0						
Zr	2	4	504	-214	-463	18	16						
Dy	0	-0	-46	9	33	2	2						
Sc	0	1	158	-18	27	4	4						
Y	1	-1	-228	70	185	9	9						
Al	-0	1	-45	-1	-50	1	1						
Mn	-8	-62	23530	4193	7733	919	379						
Sr	-1	13	-1565	1196	1566	3	16						
La	0	3	-727	75	199	10	10						
Ce	2	6	-799	212	582	28	29						
Ba	4	437	-11027	10446	18771	1	166						
Li	5	10	2560	40	540	25	30						
K	-0	0	-148	5	-20	-0	-0						
Ars	16	88	4341	209	1777	26	55						
Fe	1												
Mg	0	0											
Cd	-0	0	0										

Ti	308	35	-69	420328			
V	10	1	-1	5584	379		
Be	0	0	-0	86	2	0	
Ca	-0	0	0	-66	-2	-0	0
Nb	2	0	-0	1469	20	1	-0
Cu	10	0	7	88	242	5	5
Na	-0	0	0	-51	-2	-0	0
Zr	5	1	-2	6661	92	2	-1
Dy	0	0	0	284	5	0	-0
Sc	1	0	-0	839	17	0	-0
Y	1	0	0	1345	21	1	-0
Al	0	0	-0	133	2	0	-0
Mn	113	13	50	62428	516	44	3
Sr	-5	-0	5	-5085	-124	0	5
La	1	0	0	1160	25	1	-0
Ce	4	1	0	3568	67	3	-0
Ba	-18	-1	24	-45781	-630	1	17
Li	7	1	-0	4822	95	3	-1
K	-0	0	-0	-88	-2	-0	0
Ars	11	-0	-1	1978	133	5	-1
Nb	8						
Cu	-5	2190					
Na	-0	2	0				
Zr	27	-57	-1	164			
Dy	1	1	0	5	1		
Sc	3	-4	-0	15	1	3	
Y	7	14	0	26	3	5	16
Al	0	-1	0	3	-0	1	-0
Mn	265	560	-5	1198	134	246	695
Sr	-20	354	4	-127	0	-10	5
La	7	27	0	26	4	5	18
Ce	20	34	0	73	8	12	40
Ba	-157	2110	23	-740	-17	-86	-57
Li	22	8	-1	91	5	14	25
K	-0	-0	0	-1	-0	-0	-0
Ars	19	324	-2	33	3	6	17
Al	1						
Mn	16	101129					
Sr	1	-166	418				
La	-0	765	15	25			
Ce	-0	2038	-1	50	121		
Ba	-5	-2524	1531	23	-143	16285	
Li	3	1274	-58	24	71	-527	96
K	0	-6	2	0	0	21	-1
Ars	-0	290	-31	16	49	-72	71
K	0						
Ars	-2	331					

Covariance matrix for Group C

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V
Mo	6											
Cr	48	444										
P	2914	23981	1955877									
Zn	679	6183	405341	123080								
Pb	1640	14417	994184	252758	630632							
Co	10	88	5122	1269	3194	32						
Ni	27	233	12658	3334	7536	51	145					
Fe	3	27	1807	389	1022	6	16					
Mg	-0	-1	-89	-19	-39	-0	-1					
Cd	1	12	838	233	481	2	6					
Ti	-349	-2986	-338566	-78070	-136021	-761	-1764					

V	24	210	13499	3331	7764	51	142
Be	1	10	593	147	374	3	6
Ca	0	3	208	53	93	0	2
Nb	0	3	-409	-68	30	-1	3
Cu	121	1105	70223	17603	42366	247	580
Na	0	1	-44	23	39	0	0
Zr	-5	-37	-5701	-1224	-2910	-9	-20
Dy	-0	-3	-510	-75	-123	-1	-2
Sc	-0	0	-584	-105	-45	1	0
Y	-1	-2	-1748	-207	-203	1	-2
Al	0	3	247	27	111	1	2
Mn	31	214	45146	-1148	24222	1021	-109
Sr	35	283	12591	4108	9461	62	178
La	1	11	-1156	-67	146	4	7
Ce	-1	-5	-3844	-596	-591	6	-4
Ba	352	3162	213234	54185	116814	609	1465
Li	9	68	2836	687	2321	18	46
K	-0	-3	-219	-43	-103	-0	-2
Ars	73	602	37069	8475	21105	137	342
Fe	2						
Mg	-0	0					
Cd	1	-0	1				
Ti	-140	18	-184	302431			
V	18	-1	6	-755	252		
Be	1	-0	0	-33	7	0	
Ca	0	-0	0	-132	0	0	0
Nb	1	0	-0	814	7	0	-0
Cu	74	-3	36	-8534	596	27	7
Na	0	0	0	40	0	0	-0
Zr	-2	0	-3	4083	19	-1	-3
Dy	-0	0	-0	291	-1	0	-0
Sc	0	0	-0	546	5	0	-0
Y	0	0	-1	1256	5	1	-1
Al	0	-0	0	-63	5	0	-0
Mn	23	7	-9	5455	-202	10	-23
Sr	21	-0	6	-1778	130	10	2
La	1	0	-1	1159	8	1	-1
Ce	0	0	-2	2877	-4	1	-2
Ba	153	-9	112	-58571	1256	51	28
Li	7	-0	1	1059	75	3	-0
K	-0	0	-0	47	-1	-0	-0
Ars	46	-2	16	-4143	356	18	4
Nb	4						
Cu	1	3318					
Na	0	2	0				
Zr	14	-178	1	137			
Dy	1	-12	0	5	1		
Sc	2	-5	0	11	1	2	
Y	4	-29	1	24	3	6	16
Al	0	8	-0	1	-0	0	-1
Mn	-27	1109	-5	572	20	89	142
Sr	3	626	2	-27	3	10	25
La	4	8	1	21	3	6	15
Ce	9	-51	1	52	6	12	27
Ba	-170	9013	-1	-1212	-78	-122	-314
Li	7	152	0	28	3	7	16
K	0	-7	0	1	0	0	0
Ars	5	1631	-0	-72	-6	2	-7
Al	1						
Mn	-137	130163					
Sr	2	-1343	489				
La	-1	137	47	17			
Ce	-0	651	60	31	70		

Ba	6	-3833	1026	-307	-697	137922	
Li	2	29	84	17	28	-142	45
K	0	8	-1	0	1	-24	-0
Ars	5	190	465	19	-9	3464	131
K	0						
Ars	-4	1033					

Covariance matrix for Group P

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V
Mo	4											
Cr	39	1242										
P	1792	22741	2040470									
Zn	476	7942	235686	116362								
Pb	1066	40838	453474	323259	2033867							
Co	5	66	3197	606	1538	13						
Ni	19	407	10352	3348	13336	36	171					
Fe	2	37	1539	346	800	4	15					
Mg	-0	-0	-63	-5	5	-0	-0					
Cd	1	8	272	244	627	1	4					
Ti	-1	3750	-263154	-12502	224056	324	1229					
V	36	1039	8056	6905	43612	54	383					
Be	1	6	320	57	108	2	4					
Ca	0	1	276	59	104	0	1					
Nb	1	14	114	9	247	4	9					
Cu	68	1093	62188	11695	19219	128	437					
Na	0	2	-107	49	98	0	1					
Zr	-5	-118	-7866	-1708	-3535	1	-35					
Dy	-0	-3	-225	-75	-119	1	-0					
Sc	0	-12	-1003	-134	-242	1	-2					
Y	-0	-14	-786	-272	-539	5	3					
Al	-0	-5	-627	-75	-81	-0	-1					
Mn	66	1889	106427	10952	55846	404	920					
Sr	30	163	8406	4900	9172	58	189					
La	1	-12	-1533	-124	-344	5	5					
Ce	1	-18	-2387	-459	-1160	12	13					
Ba	171	4160	165758	77508	249814	303	1883					
Li	5	2	303	-78	345	18	30					
K	-0	-2	-249	-12	-17	-0	-1					
Ars	31	193	25220	2948	1378	54	131					
Fe	2											
Mg	-0	0										
Cd	0	-0	1									
Ti	45	19	-77	319816								
V	25	0	9	11178	1328							
Be	0	-0	-0	63	7	0						
Ca	0	-0	0	-100	-1	0	0					
Nb	2	0	-1	843	27	1	-0					
Cu	70	-1	24	-616	529	15	6					
Na	0	0	0	37	3	0	-0					
Zr	-2	1	-5	4144	4	1	-2					
Dy	-0	0	-0	164	1	0	-0					
Sc	-0	0	-0	639	13	0	-0					
Y	0	0	-1	749	5	1	-0					
Al	-0	0	-0	300	6	0	-0					
Mn	134	-0	5	23197	1238	47	3					
Sr	13	-0	10	1768	350	9	6					
La	-0	0	-1	1086	32	1	-0					
Ce	1	0	-2	2484	55	2	-1					
Ba	127	-4	208	-10385	3825	12	79					
Li	4	0	-2	2748	91	3	-0					
K	-0	0	0	54	1	-0	-0					

Ars	24	-1	2	-3913	43	7	3
Nb	6						
Cu	18	5918					
Na	0	5	0				
Zr	20	-234	0	145			
Dy	1	-2	0	6	0		
Sc	2	-20	0	16	1	3	
Y	6	4	0	28	2	3	11
Al	1	-21	0	7	0	1	1
Mn	246	2742	11	622	65	20	345
Sr	11	324	2	-39	2	11	26
La	5	0	0	29	2	5	9
Ce	15	-17	0	77	4	10	22
Ba	-134	6806	34	-1248	-40	-198	-120
Li	17	2	0	79	4	12	23
K	0	-5	0	1	0	0	0
Ars	12	730	-2	-65	-0	-5	6
Al	1						
Mn	12	40039					
Sr	1	222	873				
La	2	138	41	13			
Ce	4	768	42	24	62		
Ba	-94	13562	4685	-206	-575	157005	
Li	4	781	102	25	59	-329	79
K	0	-7	-1	0	1	-22	0
Ars	-7	1144	222	-4	3	542	57
K	0						
Ars	-4	453					

Covariance matrix for Group R

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V
Mo	1											
Cr	7	3410										
P	162	-190	475966									
Zn	106	7130	32637	53824								
Pb	133	811	53444	29782	78961							
Co	1	-6	153	60	126	11						
Ni	3	97	457	516	575	7	38					
Fe	0	-1	162	27	51	1	1					
Mg	0	0	-17	3	-1	0	0					
Cd	0	3	66	80	67	0	1					
Ti	-115	-10496	-2165	-37321	-19225	427	-1117					
V	5	24	5260	560	1444	21	46					
Be	0	-2	41	7	18	1	1					
Ca	0	0	-41	31	5	-0	0					
Nb	-0	-38	242	-101	-48	2	-3					
Cu	41	590	-720	4298	4638	9	239					
Na	0	4	-74	41	22	-0	0					
Zr	-0	-137	-316	-645	-354	12	1					
Dy	0	-7	-33	-3	-15	1	0					
Sc	-0	-13	96	-56	-50	3	0					
Y	1	-28	-148	50	-44	4	1					
Al	-0	-4	-41	-12	-16	0	-0					
Mn	9	-805	8725	-2808	-8194	469	-67					
Sr	-4	49	-9776	438	411	-3	61					
La	1	-31	-27	32	-57	6	3					
Ce	1	-67	-160	113	-308	14	3					
Ba	75	697	8559	15895	29356	48	404					
Li	0	-98	1514	-313	-116	10	2					
K	0	1	-39	15	13	0	1					
Ars	-3	-38	927	-99	463	13	26					

Fe	0						
Mg	0	0					
Cd	0	0	0				
Ti	133	-5	0	397155			
V	6	-0	2	1516	270		
Be	0	0	0	65	2	0	
Ca	-0	0	0	-175	-4	-0	0
Nb	1	-0	0	1263	8	0	-1
Cu	8	1	12	-8362	268	2	1
Na	0	0	0	9	-1	0	0
Zr	4	0	-0	5825	51	2	-4
Dy	0	0	0	276	1	0	-0
Sc	0	0	0	613	9	0	-0
Y	1	0	0	1321	6	1	-0
Al	0	0	0	257	2	0	-0
Mn	32	2	8	46805	1013	27	-43
Sr	-7	2	0	-6034	-200	-1	9
La	1	0	1	1438	13	1	-1
Ce	2	0	1	3552	22	2	-2
Ba	22	2	38	-11571	488	10	9
Li	3	0	0	2782	50	1	-2
K	0	0	0	-7	0	0	-0
Ars	1	0	-0	-568	31	1	0
Nb	7						
Cu	-38	3846					
Na	-0	7	0				
Zr	26	-53	-0	162			
Dy	1	-0	0	5	1		
Sc	2	-9	0	12	1	2	
Y	6	8	1	26	3	3	13
Al	1	-2	0	5	0	1	1
Mn	231	-1351	-9	915	66	154	325
Sr	-38	222	5	-121	-3	-11	-8
La	7	1	0	28	3	4	13
Ce	18	-24	1	75	6	8	31
Ba	-59	4183	25	-227	7	-19	63
Li	14	-68	-1	62	3	7	14
K	-0	6	0	0	0	0	0
Ars	0	-133	-1	11	-1	2	-6
Al	0						
Mn	43	43334					
Sr	-2	-2695	1066				
La	1	432	-14	15			
Ce	4	1089	-48	34	92		
Ba	1	-3339	1340	43	-2	17338	
Li	2	665	-109	16	39	-170	44
K	0	3	3	0	1	16	-0
Ars	-0	-276	167	-3	-8	82	11
K	0						
Ars	-0	138					

Appendix 4

Covariance Matrices for Suburban Pollution Sources

Pooled Covariance Matrix

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V	Be
Mo	16												
Cr	23	417											
P	-97	1985	591650										
Zn	38	1000	32781	16279									
Pb	67	954	58137	16453	44515								
Co	12	43	532	185	219	32							
Ni	57	134	818	289	451	61	243						
Fe	8	17	310	32	69	8	32						
Mg	-0	1	13	3	1	0	0						
Cd	-0	1	40	17	32	0	-0						
Ti	-626	3330	243329	9200	14860	1214	-124						
V	19	172	8843	784	865	63	130						
Be	2	3	-28	13	24	2	6						
Ca	0	0	-7	4	1	-0	0						
Nb	-0	2	843	55	172	4	3						
Cu	683	1405	-8429	2212	3736	539	2660						
Na	-1	-1	-220	-9	-18	-1	-3						
Zr	-10	121	923	101	-136	33	-4						
Dy	1	2	-86	10	13	3	4						
Sc	0	14	542	46	40	7	7						
Y	6	18	-564	67	60	16	27						
Al	-1	0	-96	-13	-25	-0	-2						
Mn	173	2921	13987	17270	12173	1808	1374						
Sr	81	46	-14440	-994	-1234	6	255						
La	7	15	-1309	85	47	19	32						
Ce	12	39	-1448	228	282	42	62						
Ba	109	128	-64306	153	6973	-158	135						
Li	18	63	3252	392	578	47	94						
K	-1	-1	-179	-9	-16	-1	-2						
Ars	164	190	179	139	704	129	567						
Fe	5												
Mg	-0	0											
Cd	-0	0	0										
Ti	121	66	10	674648									
V	22	1	1	9680	379								
Be	1	0	0	-73	2	0							
Ca	-0	0	0	-20	-1	0	0						
Nb	1	0	0	1382	23	0	-0						
Cu	367	-4	-6	-24941	838	69	5						
Na	-1	0	-0	-83	-6	-0	0						
Zr	0	2	-0	6788	109	-0	-1						
Dy	0	0	0	149	3	0	0						
Sc	1	0	0	1339	29	0	-0						
Y	3	0	0	601	13	2	0						
Al	-0	0	-0	145	-1	-0	-0						
Mn	192	26	39	41725	2251	103	-4						
Sr	26	-1	-3	-9014	-265	6	5						
La	2	0	0	-1106	-2	3	0						
Ce	7	1	0	834	33	4	0						
Ba	-24	-5	3	-68785	-1568	13	11						
Li	16	1	0	4309	149	3	-1						
K	-0	0	-0	-132	-5	-0	0						
Ars	90	-1	-2	-3952	236	17	1						
Nb	8												
Cu	-9	33043											

Na	-1	-24	0				
Zr	21	-286	-3	443			
Dy	1	32	-0	5	1		
Sc	3	13	-0	20	1	4	
Y	3	232	-0	27	4	5	22
Al	0	-26	0	4	0	0	0
Mn	172	5132	-38	1516	180	373	1127
Sr	-46	3747	10	-289	-1	-28	9
La	-1	291	-0	16	5	3	28
Ce	9	538	-0	68	9	11	44
Ba	-192	5484	50	-990	-26	-179	-65
Li	17	795	-4	93	5	17	24
K	-0	-22	0	-0	-0	-0	-0
Ars	6	6820	-7	-98	8	7	58
Al	1						
Mn	-9	324714					
Sr	-1	-3473	1492				
La	-0	1493	14	47			
Ce	1	2676	7	62	117		
Ba	4	-10585	4053	114	-63	25841	
Li	0	2009	-130	20	64	-959	122
K	0	-16	5	0	0	42	-3
Ars	-7	736	828	67	137	794	217
K	0						
Ars	-6	1867					

Covariance matrix for Group A

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V	Be
Mo	20												
Cr	23	162											
P	-167	4253	642030										
Zn	26	676	38396	17093									
Pb	51	877	56863	14767	41193								
Co	15	57	477	202	214	36							
Ni	72	127	900	232	357	75	297						
Fe	11	19	303	34	77	10	41						
Mg	-0	1	20	3	1	0	-0						
Cd	-0	1	31	15	28	0	-1						
Ti	-752	4910	284886	15224	21242	1477	-353						
V	24	207	9515	971	988	72	155						
Be	2	3	-54	12	19	3	8						
Ca	0	-0	-0	5	2	0	0						
Nb	0	10	863	63	170	4	4						
Cu	863	1049	-6008	1365	3832	701	3336						
Na	-1	-3	-224	-11	-17	-1	-4						
Zr	-15	71	1708	67	5	44	-6						
Dy	1	4	-100	8	8	4	5						
Sc	0	18	637	58	56	8	8						
Y	7	23	-652	60	36	20	33						
Al	-1	-1	-122	-16	-23	-0	-3						
Mn	230	3840	2075	19763	11046	2082	1624						
Sr	104	-40	-15564	-1174	-1082	21	317						
La	9	21	-1691	62	-26	23	39						
Ce	15	51	-1720	202	197	47	75						
Ba	128	-444	-76435	-1470	7395	-202	73						
Li	23	99	3419	443	575	56	118						
K	-1	-2	-195	-12	-18	-1	-3						
Ars	210	250	-468	66	524	162	723						

Fe	6		
Mg	-0	0	
Cd	-0	0	0

Ti	175	75	12	759516			
V	26	1	0	11406	417		
Be	1	-0	-0	-96	3	0	
Ca	0	0	0	-34	-1	0	0
Nb	2	0	0	1627	28	0	-0
Cu	466	-6	-10	-29250	1122	88	6
Na	-1	-0	-0	-140	-7	-0	0
Zr	-0	2	-0	8966	152	-0	-1
Dy	1	0	0	179	4	0	0
Sc	2	0	0	1583	34	0	-0
Y	4	0	0	679	20	2	0
Al	-0	0	-0	172	-1	-0	-0
Mn	219	32	47	51978	2527	120	-1
Sr	39	-2	-3	-13942	-295	9	4
La	3	0	0	-1376	-2	3	0
Ce	9	1	0	999	44	5	0
Ba	-22	-8	2	-89514	-1931	16	10
Li	20	1	0	5736	181	3	-0
K	-1	0	-0	-167	-6	-0	0
Ars	115	-1	-3	-5142	296	21	1

Nb	9						
Cu	-1	40688					
Na	-1	-33	0				
Zr	26	-570	-3	369			
Dy	1	41	-0	6	1		
Sc	3	20	-0	26	1	5	
Y	3	290	-0	30	5	6	27
Al	0	-36	0	4	0	0	0
Mn	127	6932	-37	1972	220	437	1418
Sr	-49	4647	9	-356	-2	-36	6
La	-2	367	-0	16	7	4	34
Ce	7	672	-1	78	10	13	53
Ba	-221	5820	55	-1328	-35	-231	-109
Li	19	1053	-4	117	6	21	31
K	-0	-31	0	-1	-0	-0	-0
Ars	4	8765	-8	-123	10	10	73

Al	1						
Mn	-27	398213					
Sr	-2	-3493	1600				
La	-0	1819	25	57			
Ce	1	3104	21	74	132		
Ba	7	-12879	4535	135	-99	30319	
Li	0	2254	-111	23	75	-1115	139
K	0	-18	5	0	0	48	-3
Ars	-8	765	1089	83	169	1045	269
K	0						
Ars	-7	2381					

Covariance matrix for Group B

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V
Mo	3											
Cr	13	145										
P	1219	9782	750758									
Zn	123	884	70704	10610								
Pb	297	1922	161719	23547	53047							
Co	-3	16	23	50	-17	28						
Ni	2	41	2292	357	705	16	25					
Fe	1	16	1061	37	88	0	2					

Mg	-0	-0	-43	2	1	1	0
Cd	0	3	279	39	89	-0	1
Ti	-288	-3316	-230290	-1217	-5755	92	13
V	16	179	11815	978	2121	18	49
Be	0	1	56	13	25	1	1
Ca	0	-1	-47	7	17	-0	-0
Nb	-1	10	53	54	53	9	11
Cu	18	148	10705	1598	3439	28	75
Na	-1	-7	-514	-22	-54	0	-1
Zr	2	38	1717	472	913	17	34
Dy	-1	-5	-415	-31	-74	1	-1
Sc	-1	-4	-479	-20	-69	4	1
Y	-3	-33	-2264	-96	-227	-0	-5
Al	-0	2	-16	-18	-51	2	1
Mn	-188	1482	30411	-4383	-16480	1654	678
Sr	-25	-373	-22761	-720	-1530	-35	-64
La	-3	-17	-1507	-39	-130	4	3
Ce	-14	-39	-5571	-329	-1029	45	22
Ba	6	-371	-13776	4911	10847	-5	79
Li	3	98	5068	147	221	20	24
K	-0	-2	-205	-7	-21	1	0
Ars	7	53	4290	458	1048	0	12
Fe	3						
Mg	-0	0					
Cd	0	-0	0				
Ti	-673	42	-26	180471			
V	21	-0	4	-4454	224		
Be	-0	0	0	22	1	0	
Ca	-0	0	0	78	-2	0	0
Nb	0	0	-0	212	13	0	-0
Cu	5	1	5	-48	169	3	1
Na	-1	0	-0	297	-10	0	0
Zr	-1	1	1	1114	44	1	0
Dy	-1	0	-0	168	-7	-0	0
Sc	-1	0	-0	242	-5	0	0
Y	-5	0	-0	1316	-43	0	1
Al	0	0	-0	-102	3	0	-0
Mn	231	14	-23	-58895	1915	29	-56
Sr	-61	2	-3	14807	-495	-1	8
La	-4	0	-0	1019	-22	0	0
Ce	-9	2	-2	2579	-47	1	-0
Ba	-130	7	17	36885	-624	8	23
Li	15	-0	1	-3492	132	1	-2
K	-0	0	-0	124	-3	0	0
Ars	5	-0	2	-1084	62	0	-0
Nb	7						
Cu	21	290					
Na	0	-4	1				
Zr	18	101	0	59			
Dy	0	-4	0	-0	0		
Sc	2	-1	0	3	0	1	
Y	-0	-16	2	1	1	2	11
Al	1	-1	-0	0	0	0	-1
Mn	427	380	-82	186	1	194	-421
Sr	-21	-157	27	-10	15	14	121
La	4	2	2	11	1	2	7
Ce	24	0	5	46	4	11	18
Ba	-9	611	54	277	19	33	247
Li	13	42	-6	18	-3	-1	-30
K	0	-0	0	1	0	0	1
Ars	-1	62	-3	8	-2	-2	-11
Al	0						
Mn	179	129188					

Sr	-12	-6767	1444				
La	-0	-126	70	7			
Ce	3	2008	118	26	139		
Ba	-38	-16564	3176	155	155	11258	
Li	4	2327	-384	-13	2	-853	114
K	0	13	9	1	3	21	-2
Ars	-0	104	-103	-9	-36	35	21
K	0						
Ars	-1	27					

Covariance matrix for Group P

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V	Be
Mo	0												
Cr	-1	433											
P	-35	6314	233977										
Zn	53	349	14274	23313									
Pb	163	1559	39811	48814	136852								
Co	-0	47	444	386	519	26							
Ni	1	133	1690	407	1282	20	48						
Fe	-0	7	246	34	64	2	2						
Mg	-0	3	21	-1	-8	0	1						
Cd	0	2	45	29	73	1	1						
Ti	-87	18814	279047	-7986	20757	512	5304						
V	-5	447	10524	-60	-244	64	122						
Be	0	0	-23	39	98	1	1						
Ca	-0	1	-47	-16	-44	-0	0						
Nb	1	7	142	232	698	5	7						
Cu	9	145	2064	2978	7990	67	97						
Na	-0	5	-16	-22	-31	-1	1						
Zr	-0	-230	-1284	-102	-502	-17	-75						
Dy	0	-2	-42	60	139	0	0						
Sc	-1	30	492	3	-107	6	8						
Y	1	-15	-282	230	511	0	-1						
Al	-0	4	4	-6	-56	1	1						
Mn	49	1818	19685	38418	57605	1255	1023						
Sr	-5	363	-4281	-3556	-6552	-119	65						
La	1	-25	-534	451	891	13	0						
Ce	3	-15	-772	1124	2680	27	17						
Ba	6	647	-12410	47	-3980	-3	221						
Li	1	-49	-83	772	1553	22	-4						
K	-0	1	-75	-14	-30	-0	0						
Ars	5	35	319	1127	3876	13	35						

Fe	0						
Mg	0	0					
Cd	0	0	0				
Ti	167	133	36	961963			
V	14	2	2	17941	682		
Be	0	-0	0	-50	-2	0	
Ca	-0	0	-0	125	-1	-0	0
Nb	1	-0	0	-198	3	1	-0
Cu	7	-0	5	992	65	6	-3
Na	-0	0	-0	372	1	-0	0
Zr	-0	-2	-0	-11494	-166	-0	-2
Dy	-0	-0	0	-158	-6	0	-0
Sc	1	0	0	1203	40	-0	0
Y	-0	-0	0	-807	-32	0	-0
Al	0	0	0	159	4	-0	0
Mn	91	14	48	24253	1801	63	-12
Sr	-21	6	-6	31846	-53	-6	14
La	1	-0	1	-2367	-30	1	-1
Ce	2	-0	2	-3183	-48	3	-1

Ba	-27	12	-5	47188	-58	2	25
Li	2	-1	1	-4725	-30	2	-2
K	-0	0	-0	57	-1	-0	0
Ars	1	-1	2	-460	-32	3	-2
Nb	4						
Cu	46	533					
Na	-0	-3	0				
Zr	0	-45	-6	189			
Dy	1	8	-0	2	0		
Sc	-1	1	0	-14	-0	3	
Y	2	27	-0	8	1	-2	4
Al	-0	-1	0	-3	-0	1	-0
Mn	300	4504	-20	-1316	64	216	246
Sr	-61	-545	27	-497	-9	12	-30
La	6	69	-1	20	1	-2	5
Ce	17	188	-2	14	3	-3	12
Ba	-58	-317	45	-1132	-10	34	-19
Li	11	119	-4	65	3	-2	9
K	-0	-1	0	-2	-0	0	-0
Ars	22	239	-1	-0	4	-5	14
Al	0						
Mn	49	95717					
Sr	5	-5068	2833				
La	-0	799	-163	17			
Ce	-1	1844	-288	35	83		
Ba	14	9668	4107	-129	-145	10723	
Li	-0	1116	-388	30	58	-535	65
K	0	-5	10	-0	-0	20	-1
Ars	-2	908	-210	28	83	-315	50
K	0						
Ars	-1	131					

Covariance matrix for Group R

	Mo	Cr	P	Zn	Pb	Co	Ni	Fe	Mg	Cd	Ti	V	Be
Mo	1												
Cr	36	2134											
P	-207	-17228	352004										
Zn	71	3360	-10554	10576									
Pb	37	818	32109	11351	25208								
Co	0	-37	1118	53	226	8							
Ni	6	214	-641	573	616	8	62						
Fe	0	11	77	20	13	0	1						
Mg	0	2	-10	4	1	0	0						
Cd	0	5	4	14	17	0	1						
Ti	-180	-10207	155300	-17911	-19678	279	-902						
V	-0	-164	2723	-141	26	19	4						
Be	0	-0	102	11	26	1	1						
Ca	0	3	-13	9	7	0	1						
Nb	-1	-54	1318	-68	24	2	-4						
Cu	76	4670	-35613	7513	1543	-95	437						
Na	0	9	-160	14	-8	-1	1						
Zr	10	611	-3478	249	-1304	-11	20						
Dy	0	0	120	18	31	1	1						
Sc	-0	-11	365	13	42	2	2						
Y	1	20	558	113	148	4	9						
Al	0	5	-2	6	-14	-0	-0						
Mn	1	-1886	80580	1685	12596	356	208						
Sr	11	632	-8058	1058	49	-17	65						
La	1	6	879	138	240	6	9						
Ce	2	12	1653	255	389	11	15						
Ba	73	3737	-28465	8571	7132	-6	515						

Li	-1	-133	2797	9	352	14	8
K	0	10	-113	15	-1	-0	1
Ars	0	-78	2574	74	436	14	15
Fe	0						
Mg	0	0					
Cd	0	0	0				
Ti	77	-8	-2	219893			
V	0	0	-0	1101	76		
Be	0	0	0	25	1	0	
Ca	0	0	0	-24	-0	0	0
Nb	1	-0	0	932	7	0	-0
Cu	30	4	12	-17984	-385	-1	7
Na	-0	0	0	-56	-2	-0	0
Zr	6	1	0	2579	-27	-0	0
Dy	0	0	0	73	1	0	0
Sc	0	0	0	286	5	0	-0
Y	1	0	0	379	7	0	0
Al	0	0	0	66	0	0	0
Mn	45	-1	9	24030	812	36	-1
Sr	0	0	1	-3679	-67	-1	1
La	1	0	0	262	13	1	0
Ce	2	0	0	692	22	1	0
Ba	7	4	11	-26158	-250	4	8
Li	1	-0	0	2003	28	1	-0
K	0	0	0	-92	-1	-0	0
Ars	1	0	1	1038	29	1	0
Nb	8						
Cu	-88	10732					
Na	-1	21	0				
Zr	4	1259	1	1167			
Dy	0	2	-0	1	0		
Sc	1	-19	-0	3	0	1	
Y	3	50	-0	22	1	1	5
Al	0	16	0	4	0	0	0
Mn	304	-4110	-39	293	43	101	255
Sr	-31	1328	5	106	-0	-6	4
La	3	16	-0	17	1	2	5
Ce	7	47	-0	35	2	3	10
Ba	-136	7623	16	699	8	-20	68
Li	10	-271	-1	-18	2	5	9
K	-0	20	0	2	-0	-0	-0
Ars	11	-163	-1	-21	2	3	9
Al	0						
Mn	4	29029					
Sr	1	-1396	293				
La	0	353	-4	8			
Ce	1	562	-8	14	30		
Ba	-1	-1767	1327	63	107	9363	
Li	0	691	-44	10	19	-184	41
K	0	-17	3	-0	-0	19	-1
Ars	-0	739	-36	10	22	-50	32
K	0						
Ars	-1	42					