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CHEMICAL COMPOSITION AND SOURCES OF AEROSOLS IN OSLO, NORWAY DURING THE WINTER 1971

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LIST OF CONTENTS

		Page
	SUMMARY	5
1	INTRODUCTION	7
2	STUDY AREA	8
3	EXPERIMENTAL	9
	3.1 Sampling and observations	9
	3.2 Neutron activation analysis	10
	3.3 Atomic absorption spectrometry	10
4	RESULTS AND DISCUSSION	11
	4.1 Aluminium	12
	4.2 Iron	13
	4.3 Chromium, manganese, zinc	13
	4.4 Vanadium	14
	4.5 Bromine, lead	14
	4.6 Cadmium	15
	4.7 Antimony	16
	4.8 Concluding remarks	16
5	REFERENCES	30
	APPENDIX I, Elemental concentration values determined for seven sets of daily air filter samples from the Oslo study area during the winter season 1970-71.	31
	APPENDIX II Matrices showing correlation coeffisients between various chemical parameters for each individual day and for the composite material.	39

SUMMARY

This work is part of an extensive study during the winter season 1970-71 of air pollution by SO_2 and black smoke in Oslo, Norway in relation to meteorological conditions. The study was based on samples of atmospheric particulates from 25 stations in the area from 6 selected days during the period 4 February - 8 March 1971. Neutron activation analysis was used for the determination of the trace elements Al, Ti, Mn, Br, Cr, Fe, Zn, Sb, and flameless atomic absorption spectrometry for Cd and Pb.

The investigation shows that the concentrations of some metallic elements could be very high during inversions in the winter season, mainly in the industrial area in the northern part of Oslo and in the lower central parts of Oslo. Some concentration values for elements such as iron, manganese and zinc are among the highest reported for urban areas in the U.S.A.

The concentrations of black smoke show high correlation with lead and bromine. This indicates that automobile traffic is a major source of black smoke in the area.

The SO_2 concentrations show the highest correlation with vanadium, bromine and lead, which indicate that besides the burning of fuel oil, automobile traffic also may be a significant source of the SO_2 pollution near the surface.

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1 INTRODUCTION

Investigations of the chemical composition of aerosols have been frequently reported in the literature. In recent years, considerable attention has been paid to trace components because of the increasing emphasis on heavy metals and other potentially toxic substances in the environment. Modern developments in analytical chemistry have facilitated the simultaneous determination of a great number of chemical elements or compounds in a single sample of atmospheric particulates.

The present paper describes an investigation of the elemental composition of aerosols by neutron activation analysis, supplemented with atomic absorption spectrometry. This work was part of an extensive study of the air quality in Oslo, Norway, during the winter season 1970-71. The main objective of the study was to measure SO_2 pollution in the area in relation to meteorological conditions (1). However, as the SO_2 pollution represents only one part of the total pollution problem, determination of a number of chemical elements collected on air filters in different meteorological situations representative for the Oslo area was included in the program. Preliminary results were presented in (1).

2 STUDY AREA

The area studied, shown in Fig. 1, consists of the city of Oslo and the Nesodden and Bærum communities, with a total population in 1970 of about 570 000. Oslo is the administrative center of Norway, and the urban area has some industry. The city is situated in a basin, at the end of the 100-km long Oslo fjord. Within a radius of 6-12 km from the city center, the area is shielded by hills of heights 200-500 meters a.s.l. The valleys with outlets into the Oslo basin are short (15-20 km). The main drainage winds are from the east-northeast (ENE), north-northwest (NNW), and south (S). The drainage of cold air along the Oslo fjord is restricted by the narrow sound at Drøbak (25 km south of Oslo) with 200-300 m high ridges on both sides.

The climate in Oslo is more continental than maritime, because the city is situated at the end of a long fjord. The average monthly temperature of January is -4.7°C. Prevailing winds during the autumn and winter are weak and mainly from the north. The air pollution levels reach a maximum during the winter with its stagnant air and inversions.

In the Oslo area, fuel oil accounts for approximately 60% of the energy consumption for heating. Thus fuel consumption in stationary sources is an important factor contributing to the air pollution. Other important sources are industrial processes, fuel consumption in mobile sources, and refuse incineration. The relative contributions from these four source categories to the emissions of the major air pollutants in the Oslo area (SO_2, particulates, CO, hydrocarbons, NO_2) have been reported elsewhere (1).

3 EXPERIMENTAL

3.1 Sampling and observations

The present study was based on samples of atmospheric particulates from 25 stations in the Oslo area. The stations are listed in Table 1 and the location of the stations are shown in Figure 2. The sampling heights were 3-10 m above the ground. Samples from 6 different days during the period 4 February to 8 March, representing different meteorological conditions (Table 2), were selected for the analysis of elemental composition. All selected days, except 7-8 March, were weekdays.

The atmospheric particulates were collected on paper filters (Whatman No.1). The air volume passing through each filter was nominally 3.6 m^3/day. 24-hours samples were collected at all stations, and the filters were changed at 14 h local time. Prior to the elemental analysis (see below), the filters were subject to determination of black smoke (SM) by reflectometry (OECD standard method 1964 (2)).

The daily mean concentrations of SO_2 were determined spectrophotometrically by the Thorin method, after absorption in an acid hydrogen peroxide solution (2).

The atmospheric stability in the Oslo area was assessed by means of recordings by termographs, situated at 6 different heights outward from the centre of Oslo (10 m a.s.l.) up to 420 m a.s.l. along the slope of the Holmenkollen hill northwest of the city. The air stability was classified as follows:

Stable : The temperature increases with height.
Unstable : The temperature decreases with height more than $1^{o}C/100$ m.
Neutral : The temperature decreases with height between 0 and $1^{o}C/100$ m.

The local wind was recorded at 5 stations (Figure 2, stations A-E).

3.2 Neutron activation analysis

The method used for multi-element analysis of the filters by neutron activation has been reported in detail elsewhere (3), and only a brief description is given here. The filters were first irradiated for 5 minutes in the JEEP-II reactor (Kjeller, Norway) at a thermal neutron flux of $1.5 \cdot 10^{12} n\ cm^{-2} s^{-1}$, and then subjected to γ-ray spectrometry using a Ge(Li) solid-state detector, for the determination of elements yielding short-lived isotopes upon neutron activation. The filters were then activated for 3 days at a neutron flux of $5 \cdot 10^{12} n\ cm^{-2} s^{-1}$. After 14 days' storage for the decay of short-lived activities, another measurement by γ-ray spectrometry was carried out in order to determine elements giving rise to long-lived isotopes upon activation. The quantitative evaluations were made possible by means of standards prepared on the same type of filters, and irradiated at the same conditions as the filter samples. The following trace elements were determined:

Short-period irradiation: Al, Ti, V, Mn, Br
Long-period " : Cr, Fe, Zn, Sb

Attempts to determine the elements Na, Cl and Ca were unsuccessful because of high filter paper blanks for these elements. For the other elements listed above, the blanks were sufficiently low and reproducible for reliable analyses. The elements Sc, Ti, Se, Ag, In, I, Cs, Ba, La, Sm, Eu, Tb, Dy, Hf, Ta, Au, Th and U were also determined in one or more of the runs, but were found to be present in amounts close to or below the analytical limit of detection in all or most samples tested.

3.3 Atomic absorption spectrometry

After completion of the activation analysis, the filter samples were analysed for Cd and Pb using flameless atomic absorption spectrometry. The filters were cut into pieces and leached with 1:1 HNO_3 in centrifuge tubes at $80^{\circ}C$. After dilution with distilled water and centrifugation of the paper mass, 20 µl samples were transferred to a graphite furnace (Perkin-Elmer HGA 72) and atomized.

The absorption was measured with a Perkin-Elmer atomic absorption spectrophotometer (Model 300) with a deuterium background corrector. Comparisons were made with the standard solutions of Pb and Cd, with approximately the same nitric acid concentration as the sample solutions.

4 RESULTS AND DISCUSSION

The analytical results for the 6 sets of 24-hours filter samples are given in Appendix I. Inter-element correlation coefficients calculated for the entire data set and for data from each day are given in Appendix II. A list of the highest correlations found is given in Table 3, where all correlation coefficients are statistical significant at a 95% or higher level. In the correlation calculations, data for the air concentration of SO_2 and SM are also included.

In Figures 3-8 the area distributions of the different trace elements are presented for 3 typical meteorological situations as follows:

a) 5-6 February 1971: Stable air with weak northerly winds (down-valley) during the night and weak southerly winds (up-valley), or stagnating air during daytime (about 8 hours).

b) 11-12 February 1971: Neutral air stability. The local wind was steady from south-westerly directions are relatively strong (2-6 m/s).

c) 24-25 February 1971: Unstable to neutral air stability. The local wind was steady from northerly directions and relatively strong (3-10 m/s).

A comparison of the analytical results from the different sets shows that very high concentrations of most of the elements were found during days with strong inversion in the area (Figures 3-8 and Table 2). The concentrations were low during the days when the air stability was neutral and unstable. However, the spatial distributions of most analysed elements differed somewhat due to the different wind conditions. At the hillside stations in the northern parts of the area the concentrations were generally higher on 11-12 February (southerly wind) than during the 24-25 February period.

Some trace element concentration data during two of these situations are given in Table 4. The data are from three stations: station 6 situated in the centre of Oslo, station 8 situated in a residental area about 1.2 km south of a steel work and a galvanizing plant, and station 11 near the city centre but situated on a hill approximately 140 m a.s.l. In the lower part of Table 4, the maximum and the 90 percentiles of the 24-hours concentrations of these elements measured in several urban areas in USA (4, 5) are listed for comparison.

In the following, the Oslo observations are discussed for each of the trace elements studied:

4.1 Aluminium

This element is one of the major components of the earth's crust, and is presumably closely associated with particulates, such as rock dust, soil particles, etc. The aluminium concentrations appear to be rather independent of the weather conditions, and are fairly uniform over the whole area, indicative of a crustal source (Figure 4). Stations 10 and 14, and to a lesser extent station 23, show a higher level of aluminium than the other stations. This may in part be explained by contributions from local cement industry (station 14) and a stone quarry (station 23) nearly. Aluminium does not appear to be appreciably correlated with any of the other elements studied, which indicates that the earth's crust component of the dust is not a significant source for any of these.

4.2 Iron

The iron concentrations and distribution in the Oslo area are very much dependent on the weather conditions. During inversion periods very high iron concentrations occurred in the lower central part of the city (Table 4). Comparison of the observed distribution patterns (Figure 4) with meteorological data clearly indicates a steel mill in the industrial area in the northern part of the city (between station 8 and 17 in Figure 2), as a major source of iron.

Iron is almost as abundant as aluminium in the lithosphere. Thus the iron content in atmospheric particulates may in general thus contain both a geological and an industrial component. The low correlation of iron with aluminium found in this study is additional evidence that iron in the Oslo air is predominantly of industrial origin.

4.3 Chromium, manganese, zinc

These elements all show high correlations with iron, and are also very well inter-correlated. They are therefore discussed as a group. The distribution patterns of these elements during inversion periods are very similar to that of iron (Figures 4, 5 and 6), pointing to the same source region, and very high concentration of manganese and especially of zinc are observed in certain areas.

The concentrations of zinc in the Oslo air can under inversion conditions be three times as high as the maximum 24-hours concentration values measured in urban areas in USA during the period 1957-1966. The most probable source of zinc is a (4,5) galvanizing plant situated in the previously mentioned (section 4.2) industrial area (Table 4). Another possible source is abration of automobile tires, which should result in a more even distribution throughout the area. However, such particles are usually in a aerodynamically large size range (6), and therefore probably poorly collected by the type of sampler used.

4.4 Vanadium

Vanadium appears not to be appreciably correlated with the metallic elements discussed above, but shows a fairly high correlation with the SO_2 content of the air in most cases. This seems to be due to the well-known fact that vanadium is present at a relatively high concentration in many fuel oils. The rather similar distribution patterns for SO_2 and V, as evident from Figures 3 and 6, give further support to the assumption that most of the vanadium comes from the burning of fuel oil. A certain fraction may, however, be associated with crustal material. The concentration levels of vanadium present in Oslo air are low compared with values measured in several US cities (4, 5).

4.5 Bromine, lead

The main source of these two elements (Figure 7) in an urban atmosphere is supposed to be automobile exhaust, because of the use of tetraethyl lead and ethylene dibromide as gasoline additives. As might be expected, the two elements are strongly correlated in the Oslo samples. The correlation of these elements with the other elements studied is considerably less pronounced. The Br/Pb ration in gasoline (with these elements as additives) is reported to be different; in U.S. 0.39 (7), in Australia 0.61 (9) and in Norway (Oslo) 0.43 in 1970 (private communications from the Norwegian Petroleum Institute). In the atmospheric particulates the ratio has been found lower by some investigators (7, 8) but small differences has also been found (9). This difference may be explained by the possibility of bromine to be lost from the particles to the gas phase after emission to the atmosphere. In the present work, the following average values for the Br/Pb ratio were observed during the selected periods:

4.2 - 5.2:	0.41
10.2 - 11.2:	0.38
11.2 - 12.2:	0.37
24.2 - 25.2:	0.42
7.3 - 8.3:	0.36

The observed mean ratios are all close to the reported value for the ratio (0.43) in gasoline in Oslo. This indicates the automobile traffic as a main source of lead in the atmospheric particulates in the area. There seems to be no evidence that bromine of marine origin contributes significantly to the measured values of this element (compare the 11.2 - 12.2 and 24.2 - 25.2 periods). Some of the daily lead concentrations recorded at stations in the centre of Oslo during inversion periods are high, but not exceptionally high when compared with US urban values (4, 5).

The correlation of lead and bromine with black smoke and SO_2 respectively indicate that automobile traffic is a major source of particulate material and also a considerable source of the SO_2-pollution at the sampling level in the Oslo atmosphere. In addition to exhaust particles, particles originating from the mechanical action between the car tires and the road surface, may contribute to the black smoke. (The annual wear of asphalt in Oslo amounts to approximately 120 000 tons).

4.6 Cadmium

The cadmium concentrations measured in the Oslo atmosphere are in most cases quite low, and do not seem to vary with the weather conditions (Figure 8). This excludes the galvanizing plant (Zn) or other industries in the northern part of the measuring area as dominant sources of cadmium, because with southerly winds (11-12 February) the emissions are transported out of the area.

4.7 Antimony

The concentration levels of this element (about 0.01 $\mu g/m^3$) are similar to these of cadmium. The antimony concentrations seem to be little affected by weather conditions (Figure 8). The correlations observed seem to affiliate antimony to some extent with industrial activity, but the correlation with bromine and lead in some cases may point to an association with automobile traffic.

4.8 Concluding remarks

The present investigation has shown that the concentrations of some metallic trace elements in the air of Oslo vary with the wind and stability conditions, and may be very high during inversions in the winter season. The high levels are mainly restricted to the industrial areas and lower central parts of the city. Some of the concentrations found for iron, manganese and zinc are among the highest reported in the literature from other urban areas (Table 4). The results of the investigation indicate that the various industrial sources in the northern part of Oslo cause most of these high concentrations.

The black smoke values (SM) show high correlation with lead and bromine. This indicates that the automobile traffic is a major source of black smoke at the sampling level.

The SO_2 concentrations are best correlated with vanadium, bromine and lead, indicating that besides the burning of fuel oil, the automobile traffic may also be a significant source of SO_2 pollution.

The possibility that the high trace metal content in the Oslo air may be a significant factor contributing to the worsening of the public health conditions, sometimes observed in Oslo during the winter season (10), cannot be excluded.

Since 1970/71, considerable efforts have been made to reduce the emission of air pollutants including trace metals in the Oslo area. It would be instructive and desirable to carry out an investigation, similar to the one described in this report, in the relatively near future, in order to ascertain the possibly beneficial effect of recent efforts to reduce air pollution in the Oslo area.

Furthermore, any future investigations should also include particle size distribution information. This would allow a more definite assessment of particulate sources and particle formation mechanisms in the Oslo urban area.

Table 1: Location and description of sampling sites

Station	Location	Height a.s.l. (m)	Description of location	Air volume (m^3/day)
1	St. Olavsplass	22	Commercial and offices	2.0
2	Haakon VII's gt.	25	"	2.4
3	Briskeby	15	Commercial-residential	2.4
4	Heimdalsgt.	11	Industrial-offices	3.6
5	Mariboes gt.	16	Light industry-residential	3.6
6	Stortorget	14	Commercial and offices	3.6
7	Kingos gt.	41	Residential-light industry	2.4
8	Sagene	86	Industrial-residential	3.6
9	Ullevål sykehus	81	Residential-offices-hospital	3.6
10	Økern	94	Industrial	2.4
11	Ekeberg	143	Residential-school	2.4
12	Sjursøya	6	Industrial	3.6
13	Malmøya	7	Residential	3.6
14	Bryn	90	Industrial	2.0
15	Nyland	125	Residential-offices	2.0
16	Østensjø	136	Residential	3.6
17	Grefsen	195	Residential	3.6
18	Kringsjå	200	Residential	3.6
19a	Huseby blindeskole	141	Residential-schools	3.6
19b	Huseby folkeskole	141	Residential-schools	3.6
20	Smestad	58	Residential	3.6
21	Skøyen	12	Industrial-offices	3.6
22	Lysaker	54	Residential	3.6
23	Sandvika	7	Industrial-residential	3.6
24	Snarøya	6	Residential	3.6
25	Nesodden	19	Residential	3.6

Table 2: The meteorological conditions during the 6 days selected for analysis of elemental composition of air filters.

Units: Air temperature in ^{o}C, cloud cover in oktas, wind direction on the scale 000-360, wind speed in m/s.

Date	Air temperature at st D 94 m a.s.l.		Air stability		Cloud cover			Geostrophic wind				Local wind at station A			
								00 GMT		12 GMT		00 h		12 h	
	00 h	12 h	00 h	12 h	19h	07h	13h	Dir	Speed	Dir	Speed	Dir	Speed	Dir	Speed
4/2-5/2	+1.8	+5.9	inversion	inversion	0	1	2	310	12	340	12	070	1.3	220	1.3
5/2-6/2	+0.8	+5.1	inversion	inversion	2	2	2	330	15	340	7	090	0.6	120	0.5
10/2-11/2	-1.8	+1.0	neutral	neutral	8	8	6	230	10	220	10	180	0.5	300	0.6
11/2-12/2	+1.9	+2.7	neutral	neutral	7	8	8	220	15	220	18	180	2.4	180	4.8
24/2-25/2	+4.4	-3.7	unstable	neutral	7	4	0	040	15	020	9	340	3.0	020	6.7
7/3-8/3	-3.3	+4.1	inversion	inversion	7	7	5	360	12	070	8	090	0.6	270	1.1

Table 3: List of the highest inter-element correlations.

Compo-nent	4/2 - 5/2	5/2 - 6/2	10/2 - 11/2	11/2 - 12/2	24/2 - 25/2	7/3 - 8/3	Composite material
SM	Pb, Br, S, Mn, Al, Fe, Cr, Sb, Cd	Br, V, S, Mn, Fe, Cr, Zn, Sb	S	Br	Br, V, Sb, Pb, S	S, V, Pb	Br, S, Mn, Fe, Cr, Zn
Br	SM, Pb, S, Sb, Al	SM, V, S, Mn, Fe, Cr Zn, Sb	Pb	Pb, SM	SM, Pb, V, Sb, S	Al	SM, Mn, Fe Cr, S
Mn	Zn, Cr, Fe, SM, S, Pb, Cd	Fe, Cr, Zn, SM, V, Sb Br, S	none	none	Fe, V, Zn Pb	Fe,Zn,Cd, Sb, Pb	Fe, Zn, Cr, SM, Br
V	Fe, Pb	SM, Br, Mn Fe, Cr, Sb, S, Zn	none	S	Fe, Pb, SM Br, Mn, Zn, Sb, S	SM, Cr, S	S
Al	Sm, Br, Sb, Pb	none	Cd, Pb	none	none	Br	none
Fe	Mn, Cr, Zn, SM, V, Cd, Pb, S	Mn, Cr, Zn, SM, V, Sb, Br, S	Zn	none	Mn, V, Zn, Sb, Pb	Mn, Sb, Zn, Cd, Pb	Mn, Cr, Zn SM, Br

Doubly underlined: $r>0.9$
Underlined : $0.9>r>0.8$
Not underlined : $0.8>r>0.6$

SM : Black suspended particulate matter
NI : Not included
S : SO_2
None: No values of $r>0.6$

Table 3: Con.

Compo-nent	4/2 - 5/2	5/2 - 6/2	10/2 - 11/2	11/2 - 12/2	24/2 - 25/2	7/3 - 8/3	Composite material
Cr	Mn, Zn, Fe, SM, Cd, Pb, S	Mn, Zn, Fe, Sb, SM, V, Br, S	none	none	none	V	Fe, Zn, Mn, SM, Br
Zn	Mn, Fe, Cr, Cd, Pb	Mn, Fe, Cr, SM, V, Sb, Br, S	Fe, Cd	none	Fe, Mn, V Pb	Mn, Cd, Fe, Sb, Pb	Mn, Fe, SM, Cr
Sb	SM, Br, Al, Cd, Pb	Cr, SM, Mn, V, Fe, Zn, Br, S	none	none	SM, Br, V, Fe, Pb, S	Mn, Fe, Zn Cd Pb	none
Cd	SM, Mn, Fe, Cr, Zn, Pb, Sb	NI	Pb, Al, Zn	none	none	Mn, Zn, Fe, Sb, Pb	NI
Pb	SM, Br, Mn, V, Al, Fe Cr, Zn, Sb Cd, S	NI	Cd, Br, Al	Br	SM, V, Fe Br, Mn, Zn, Sb, S	SM, Mn, Fe, Zn, Sb, Cd	NI
S	SM, Pb, Br Mn, Fe, Cr	SM, Br, V Cr, Mn, Fe, Zn, Sb	SM	V	SM, Br, V Sb, Pb	SM, V	SM, Br, V

Doubly underlined: r>0.9
Underlined : 0.9>r>0.8
Not underlined : 0.8>r>0.6

SM : Black suspended particulate matter
NI : Not included
S : SO_2
None: No values of r>0.6

Table 4: Daily mean concentration ($\mu g/m^3$) of some elements in suspended particulates at three stations in Oslo for two selected days. Maximum and 90 percentile 24 hr. concentrations measured in various urban areas in the U.S. (4.5) are included for comparison.

Station	Date 1971	Fe	Mn	Zn	Cd	Pb	Cr	V
6 Stortorget	5/2 - 6/2	23.0	3.05	13.8	0.013^x	3.28^x	0.092	0.051
	11/2 - 12/2	1.1	0.04	0.22	0.006	1.42	0.014	0.021
8 Sagene	5/2 - 6/2	33.9	3.64	15.9	0.011^x	2.00^x	0.089	0.029
	11/2 - 12/2	0.8	0.04	0.44	0.004	0.39	0.008	0.021
11 Ekeberg	5/2 - 6/2	0.2	0.02	0.17	-	-	< 0.004	0.003
	11/2 - 12/2	1.7	0.03	0.23	0.002	0.10	0.005	0.010
U.S. values	Site	Johnstown Penn.	Johnstown Penn.	Portland Oregon	New Rochelle N.Y.	New Rochelle N.Y.	Rochester N.Y.	New York City N.Y.
	Year	1963	1963	1964	1960	1960	1960	1957
	Max. value	16.0	6.90	4.80	0.160	16.0	0.290	2.00
	90 percen-tile	4.1	2.38	3.88	0.040	3.7	0.095	1.18

^{x}4/2 - 5/2

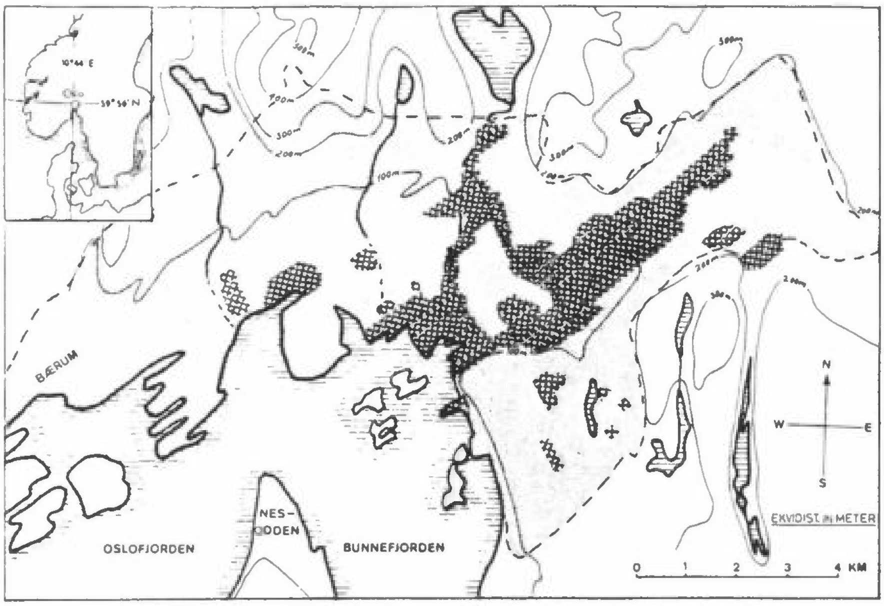

Figure 1: Oslo study area.

Residential area

Industrial area

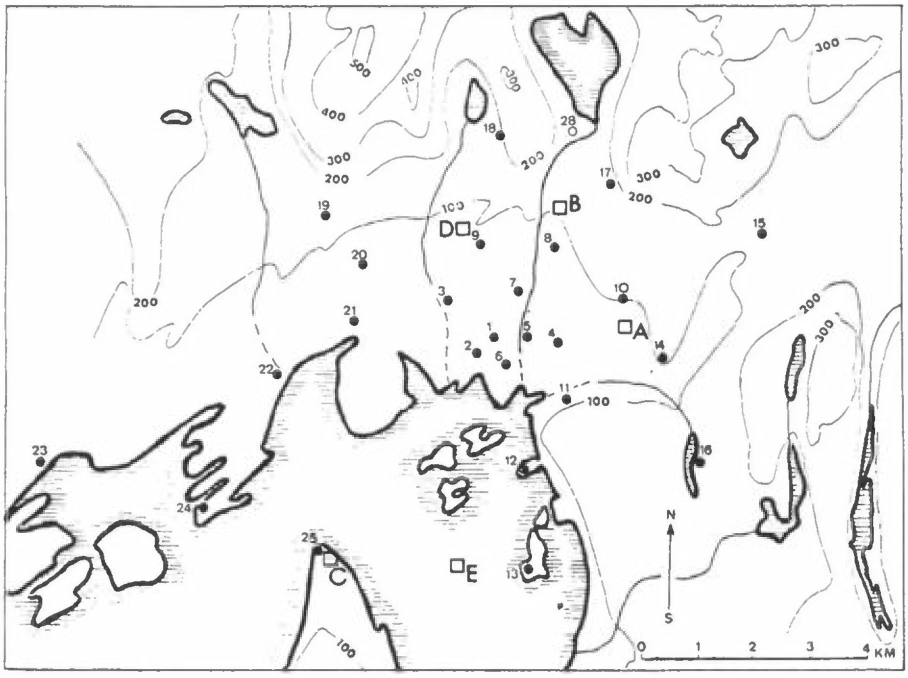

Figure 2: Sampling network in the Oslo area.

• *SO_2-stations . See also Table 1.*

□ *Wind stations, A-E*

(D: also temperature station Blindern)

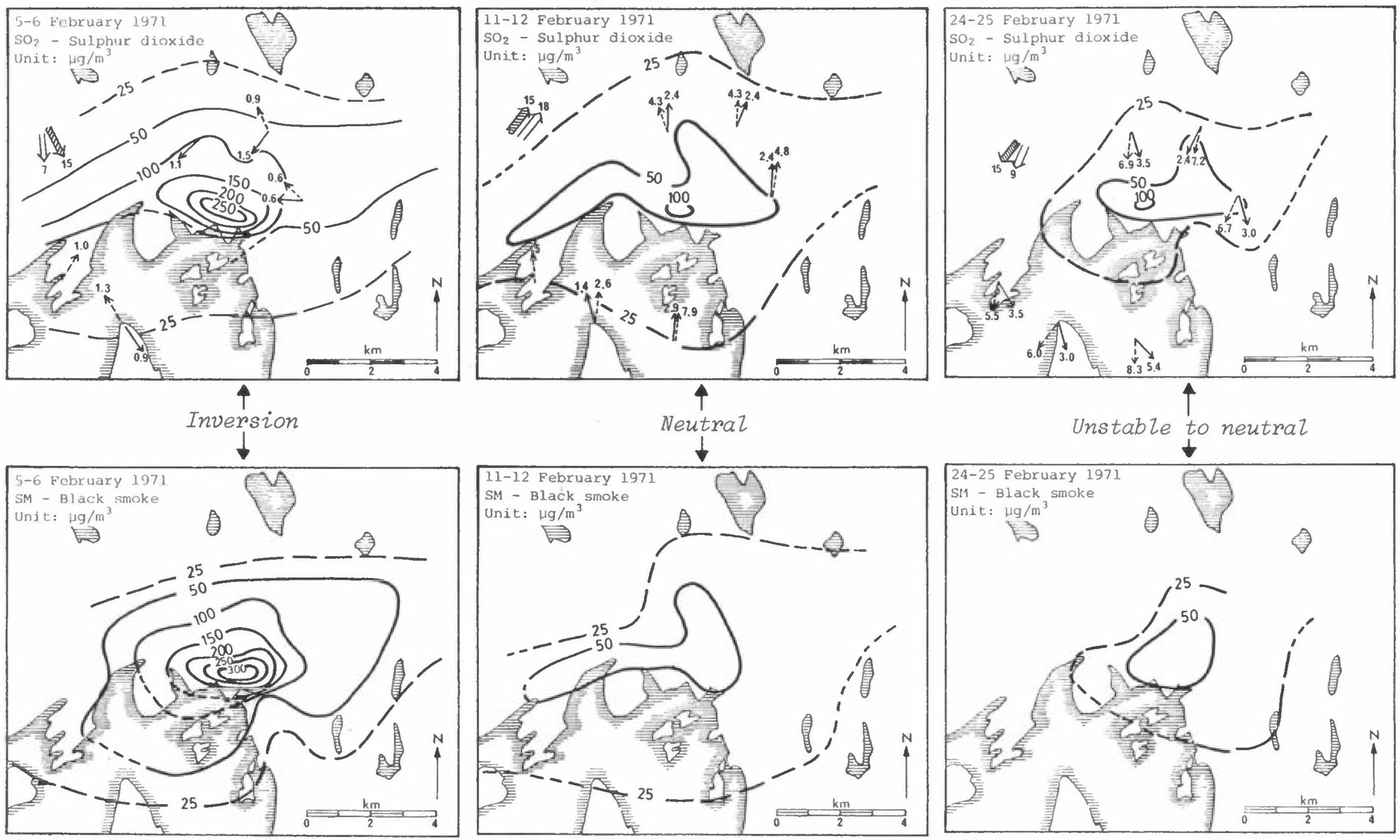

Figure 3: Distributions of SO_2 and black smoke (SM) during three days with different wind- and stability conditions in Oslo.

Geostrophic wind direction 00h ⇛ ff, 12h ⟹ ff *Local wind direction* 00h ⟶ ff, 12h --→ ff

ff: Wind speed in m/s.

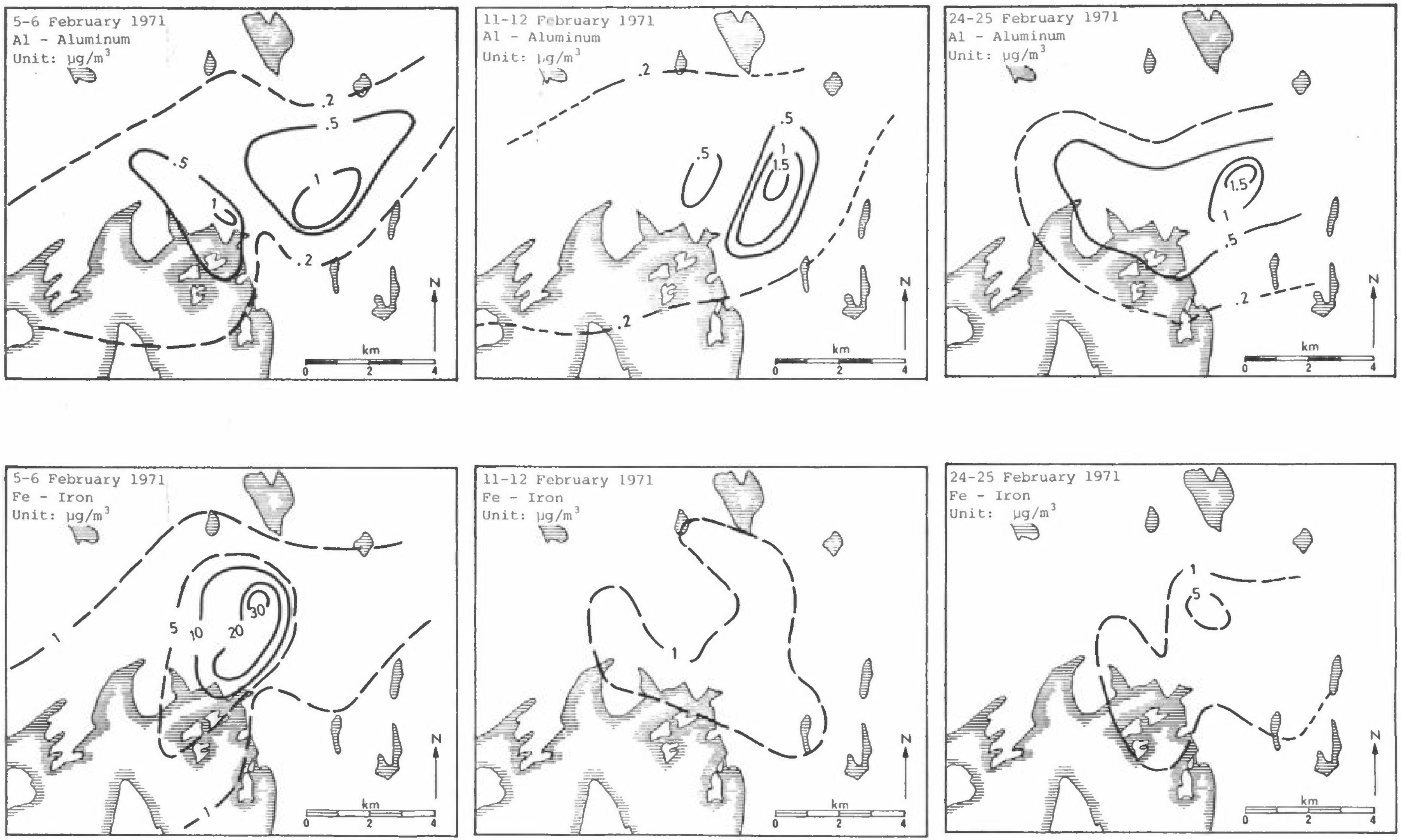

Figure 4: Distributions of aluminum (Al) and iron (Fe) during three days with different wind- and stability conditions in Oslo. Meteorological data: see figure 3.

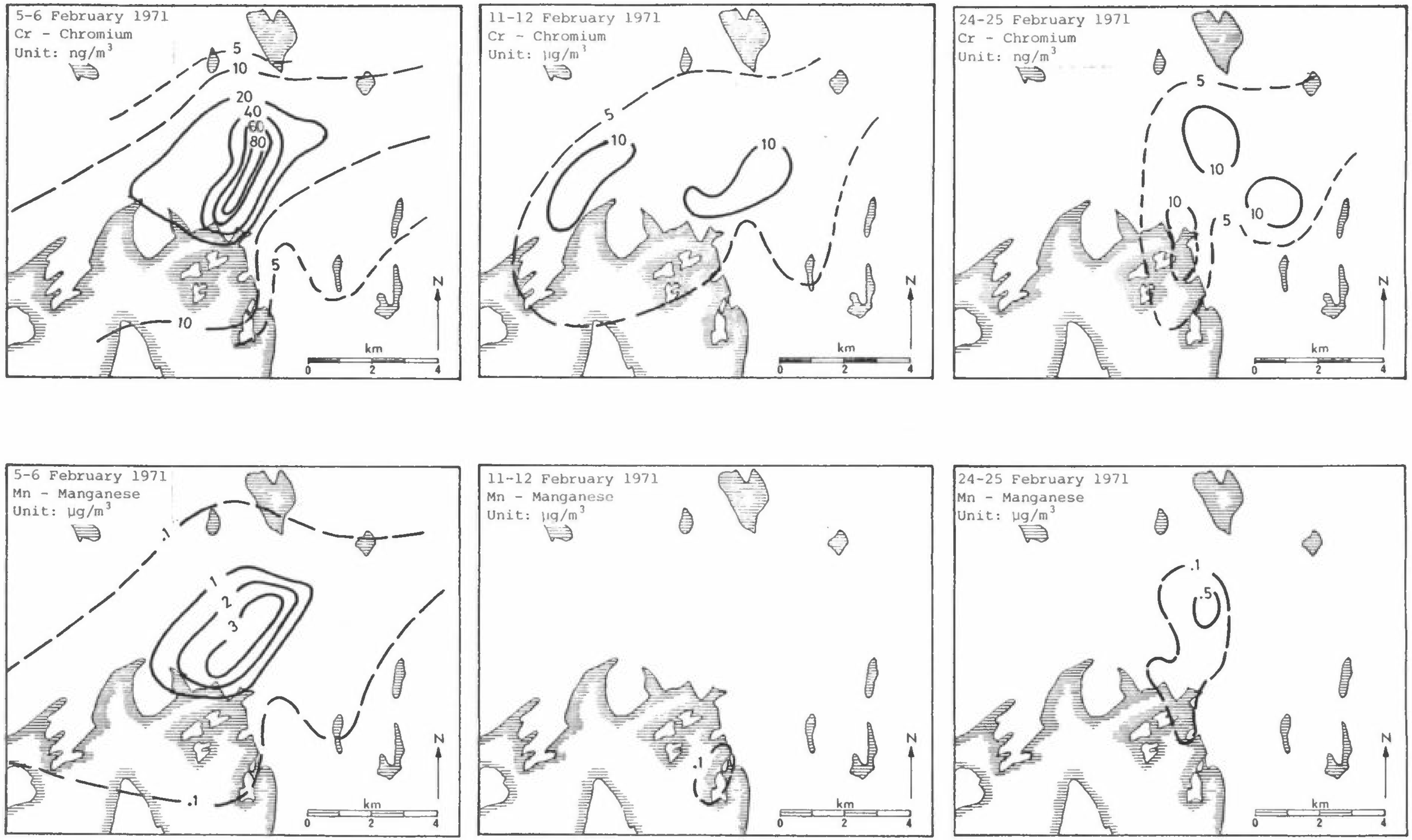

Figure 5: Distributions of chromium (Cr) and manganese (Mn) during three days with different wind- and stability conditions in Oslo. Meteorological data: see figure 3.

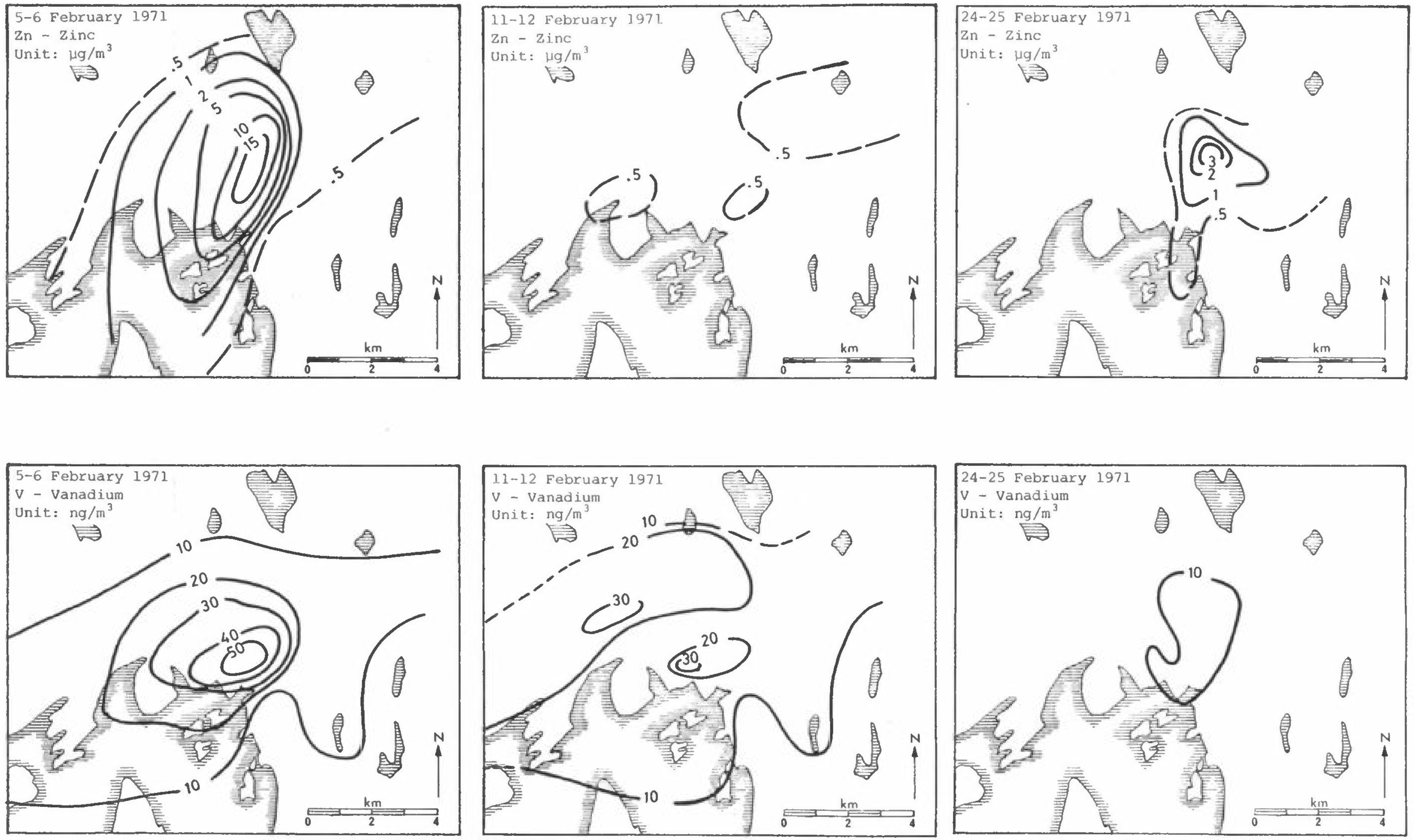

Figure 6: Distributions of zinc (Zn) and vanadium (V) during three days with different wind- and stability conditions in Oslo. Meteorological data: see figure 3.

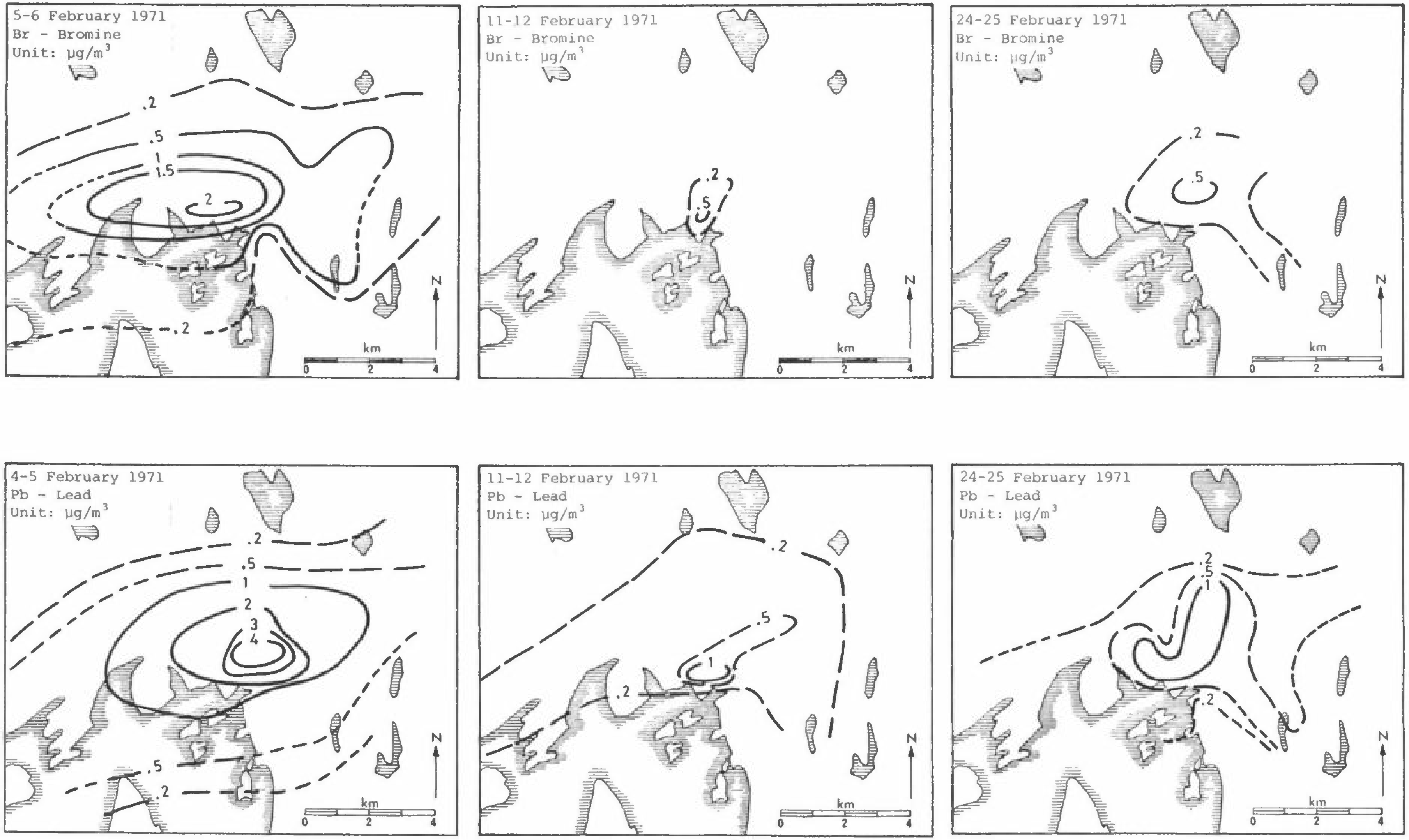

Figure 7: Distributions of bromine (Br) and lead (Pb) during three days with different wind- and stability conditions in Oslo. Meteorological data: see figure 3.

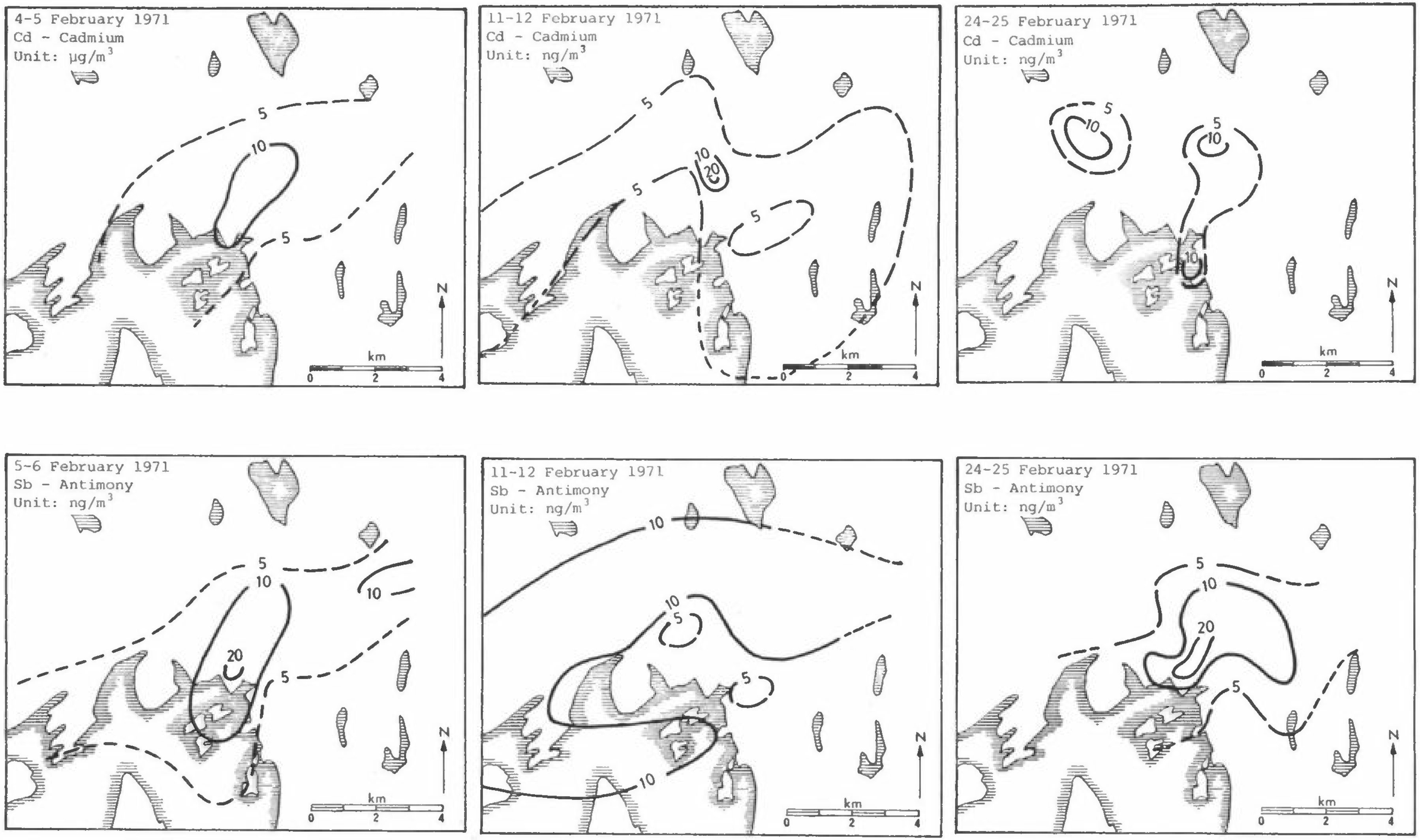

Figure 8 : Distributions of cadmium (Cd) and antimony (Sb) during three days with different wind- and stability conditions. Meteorological data: see figure 3.

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APPENDIX I

Appendix I. Elemental concentration values determined for seven sets of daily air filter samples from the Oslo study area during the winter season 1970 - 1971. All values are in μg/m^3.

4/2 - 5/2

Station	Br	Mn	V	Al	Fe	Cr	Zn	Sb	Cd	Pb	SM	SO_2
1	0.69	0.90	0.022	0.81	6.9	0.033	3.11	0.0125	0.009	2.22	195	206
2											190	195
3											140	151
4	1.78	0.30	0.055	3.78	7.2	0.025	1.31	0.0297	0.010	4.44	316	176
5	1.31	1.96	0.053	0.56	18.3	0.069	9.06	0.0261	0.019	4.33	257	171
6	1.22	0.94	0.036	0.92	6.7	0.025	2.58	0.0261	0.013	3.28	294	193
7											112	82
8	0.42	1.36	0.042	0.53	15.0	0.047	5.39	0.0200	0.011	2.00	107	92
9	0.36	0.49	0.028	0.36	3.7	0.014	2.17	0.0075	0.007	1.50	60	95
10	0.65	0.15	0.021	1.62	2.6	0.009	0.74	0.0324	0.009	1.39	87	60
11											32	36
12	0.31	0.09	0.018	0.22	1.5	0.008	0.67	0.0150	0.006	0.83	70	49
13	0.08	0.04	0.006	0.14	0.7	0.008	0.28	0.0042	0.003	0.25	20	31
14	0.69	0.09	0.011	1.53	2.5	0.008	1.33	0.0261	0.008	1.69	116	94
15	0.44	0.12	0.015	0.47	1.6	0.014	0.75	0.0192	0.008	0.89	73	86
16	0.42	0.05	0.013	0.11	0.8	0.006	0.17	0.0025	0.001	0.47	40	34
17	0.11	0.16	0.006	0.11	2.3	0.008	1.33	0.0033	0.007	0.36	28	27
18											17	28
19a	0.06	0.01	0.005	0.14	0.3	0.003	0.44	0.0019	0.014	0.17	16	37
19b	0.44	0.05	0.013	0.22	1.3	0.006	0.47	0.0028	0.004	0.78	47	40
20	0.44	0.10	0.019	0.39	1.6	0.008	0.83	0.0067	0.004	1.00	55	90
21	0.94	0.13	0.023	0.33	3.5	0.022	0.64	0.0131	0.008	1.94	101	101
22	0.22	0.20	0.040	0.31	1.6	0.011	0.75	0.0047	0.004	0.64	48	121
23	0.50	0.03	0.014	0.89	1.5	0.006	0.17	0.0031	-	0.64	16	58
24	0.25	0.07	0.019	0.25	0.8	0.008	0.44	0.0036	0.003	0.56	36	35
25	0.06	0.02	0.059	0.25	2.7	0.008	0.56	0.0053	0.003	0.50	21	29

5/2 - 6/2

Station	Br	Mn	V	Al	Fe	Cr	Zn	Sb	SM	SO_2
1									199	200
2									283	255
3									140	172
4									294	159
5	1.86	3.31	0.057	0.36	24.7	0.089	15.65	0.0180	272	166
6	2.11	3.05	0.051	1.00	23.0	0.092	13.80	0.0216	408	270
7	1.46	2.81	0.037	0.25	16.4	0.040	10.70	0.0108	155	98
8	0.69	3.64	0.029	0.53	33.9	0.089	15.90	0.0194	133	103
9	0.56	1.49	0.032	0.19	11.3	0.030	7.19	0.0089	69	108
10	0.46	0.08	0.015	0.50	2.2	0.013	0.92	0.0079	78	54
11	0.04	0.02	0.003	<0.04	0.2	<0.004	0.17	0.0008	26	35
12	0.53	0.16	0.019	0.56	2.9	0.011	1.14	0.0125	67	47
13	0.17	0.09	0.008	0.22	1.0	0.011	0.75	0.0072	28	20
14	0.83	0.08	0.009	1.36	2.8	0.006	0.44	0.0075	95	66
15	0.53	0.09	0.015	0.67	1.6	0.019	0.83	0.0100	59	74
16	0.61	0.06	0.013	0.08	0.6	0.008	0.19	0.0022	34	33
17	0.25	0.23	0.011	0.22	1.9	0.014	1.00	0.0030	34	35
18	0.19	0.25	0.012	0.22	3.0	0.011	1.47	0.0036	23	32
19a	0.17	0.10	0.011	0.17	1.2	0.008	0.72	0.0017	25	61
19b	0.22	0.08	0.005	0.19	0.7	0.006	0.06	0.0011	20	26
20	0.58	0.30	0.023	0.58	3.2	0.014	1.39	0.0042	55	79
21	1.67	0.39	0.022	0.47	4.2	0.022	1.69	0.0067	114	139
22									45	122
23	0.69	0.08	0.017	1.03	2.3	0.014	0.25	0.0047	20	67
24										
25	0.22	0.30	0.016	0.36	3.3	0.011	1.78	0.0031	40	40

10/2 - 11/2

Station	Br	Mn	V	Al	Fe	Cr	Zn	Sb	Cd	Pb	SM	SO_2
1	0.39	0.11	0.023	0.75	0.6	0.008	0.17	0.0053	0.007	0.81	85	95
2											95	152
3											55	65
4	0.36	0.22	0.040	0.39	1.2	0.008	0.36	0.0169	0.007	0.94	84	107
5	0.58	0.27	0.049	0.31	1.8	0.019	0.64	0.0114	0.014	1.22	38	87
6	0.50	0.25	0.047	0.44	0.8	0.008	0.53	0.0114	0.007	1.28	114	98
7											60	74
8	0.39	0.25	0.055	0.78	3.0	0.042	0.56	0.0208	0.008	1.03	101	133
9	0.25	0.60	0.046	0.44	2.9	0.017	1.17	0.0094	0.011	0.81	64	116
10	0.42	0.19	0.042	1.76	2.0	0.005	1.48	0.0144	0.028	3.24	53	67
11											37	40
12	0.14	0.16	0.040	0.19	0.9	0.008	0.39	0.0150	0.004	0.33	57	43
13	0.06	0.12	0.034	0.28	0.6	0.011	0.39	0.0097	0.003	0.26	26	21
14	0.25	0.12	0.022	1.14	1.1	0.014	0.31	0.0108	0.004	0.44	91	65
15	0.22	0.18	0.037	0.47	1.5	0.014	0.72	0.0150	0.011	0.67	64	93
16	0.44	0.15	0.028	0.25	0.8	0.008	0.31	0.0056	0.006	1.25	55	18
17	0.17	0.15	0.030	0.33	1.5	0.008	0.33	0.0722	0.004	0.61	44	65
18	0.22	0.31	0.036	0.28	3.4	0.011	1.08	0.0125	0.007	0.50	57	76
19a	0.11	0.18	0.028	0.36	0.9	0.006	0.31	0.0053	0.003	0.28		
19b												
20												
21	0.42	0.30	0.041	0.36	2.3	0.014	0.58	0.0086	0.008	1.08	89	89
22	0.22	0.30	0.045	0.42	0.3	0.006	0.56	0.0072	0.004	0.47	48	81
23	0.31	0.26	0.041	0.56	1.4	0.008	0.58	0.0092	0.003	0.44	69	57
24	0.11	0.21	0.040	0.31	1.3	0.008	0.31	0.0058	0.004	0.44	40	39
25	0.11	0.23	0.036	0.31	1.5	0.008	0.58	0.0089	0.007	0.44	36	38

11/2 - 12/2

Station	Br	Mn	V	Al	Fe	Cr	Zn	Sb	Cd	Pb	SM	SO_2
1	0.25	0.02	0.025	0.89	0.3	0.011	0.08	0.0058	0.001	0.44	83	76
2	0.17	0.04	0.037	0.33	1.7	0.008	0.22	0.0094	0.004	0.42	48	148
3	0.03	0.02	0.009	0.42	0.3	0.003	0.14	0.0036	0.001	0.13	33	40
4	0.17	0.05	0.029	0.39	1.1	0.014	0.61	0.0094	0.007	0.44	51	80
5	0.22	0.05	0.026	0.42	1.1	0.003	0.36	0.0092	0.008	0.53	26	62
6	0.47	0.04	0.021	0.31	1.1	0.014	0.22	0.0092	0.006	1.42	89	68
7	0.13	0.03	0.013	0.75	0.8	0.004	0.25	0.0054	0.021	0.31	50	51
8	0.08	0.04	0.021	0.31	0.8	0.008	0.44	0.0100	0.004	0.39	41	47
9	0.11	0.06	0.026	0.33	0.6	0.006	0.31	0.0092	0.007	0.31	55	63
10	0.14	0.08	0.013	1.57	1.8	0.014	0.46	0.0190	0.007	0.74	25	29
11	0.05	0.03	0.010	1.05	1.7	0.005	0.23	0.0045	0.002	0.10	32	46
12	0.03	0.07	0.015	0.33	0.9	0.006	0.28	0.0100	0.006	0.14	36	46
13	0.03	0.11	0.014	0.17	0.7	0.003	0.25	0.0083	0.008	0.13	30	31
14	0.14	0.04	0.016	0.92	0.9	0.008	0.33	0.0083	0.004	0.33	35	44
15	0.11	0.04	0.018	0.39	0.3	0.006	0.67	0.0128	0.007	0.16	41	47
16	0.11	0.09	0.018	0.19	1.3	0.008	0.36	0.0094	0.008	0.25	32	13
17	0.08	0.05	0.017	0.36	1.2	0.006	0.84	0.0128	0.004	0.24	32	30
18	0.08	0.05	0.022	0.33	1.1	0.006	0.28	0.0111	0.006	0.24	40	39
19a	0.11	0.04	0.028	0.42	1.1	0.006	0.44	0.0314	0.006	0.25		
19b	0.06	0.03	0.001	0.19	0.6	0.003	0.03	0.0014	0.001	0.08		
20	0.11	0.06	0.031	0.36	1.1	0.014	0.31	0.0119	0.008	0.39		
21	0.17	0.05	0.005	0.33	1.1	0.006	0.64	0.0111	0.008	0.44	51	48
22	0.08	0.06	0.027	0.39	0.7	0.017	0.36	0.0097	0.008	0.33	55	64
23	0.25	0.10	0.028	0.44	2.4	0.011	0.42	0.0239	0.008	0.53	55	49
24	0.03	0.05	0.017	0.28	0.8	0.006	0.39	0.0106	0.006	0.17	28	19
25	0.03	0.05	0.016	0.25	0.8	0.006	0.44	0.0114	0.004	0.17	26	16

<u>24/2 - 25/2</u>

Station	Br	Mn	V	Al	Fe	Cr	Zn	Sb	Cd	Pb	SM	SO_2
1	0.28	0.04	0.005	1.06	0.9	0.003	0.19	0.0086	0.003	0.58	59	94
2	0.33	0.14	0.012	0.78	2.3	0.006	0.33	0.0189	0.004	1.08	60	107
3											22	46
4	0.50	0.11	0.012	0.64	2.2	0.003	0.58	0.0072	0.006	1.03	69	72
5	0.69	0.19	0.016	0.83	2.5	0.006	1.06	0.0208	0.004	1.69	95	69
6	0.44	0.11	0.014	0.67	2.4	0.011	0.61	0.0208	0.006	1.08	74	90
7											54	38
8	0.28	0.52	0.018	0.44	5.2	0.014	3.17	0.0136	0.013	1.67	41	53
9	0.14	0.13	0.011	0.19	0.9	0.006	0.50	0.0086	0.004	0.39	19	49
10	0.28	0.07	0.006	1.94	3.1	0.009	1.02	0.0199	0.005	0.74	36	26
11											26	21
12	0.17	0.16	0.009	0.58	2.5	0.011	0.83	0.0058	0.013	0.33	34	28
13	0.03	0.01	0.002	0.22	0.3	0.008	0.22	0.0017	0.001	0.08	9	6
14	0.14	0.06	0.003	0.86	1.2	0.014	0.61	0.0133	0.003	0.33	40	49
15												26
16	0.22	0.04	0.005	0.33	1.0	0.003	0.31	0.0050	0.001	0.53	25	14
17	0.06	0.02	0.001	0.25	0.5	0.008	0.08	0.0014	0.003	0.14	15	10
18												
19a	0.03	0.01	0.003	0.31	0.5	0.003	0.31	0.0017	0.014	0.12	14	16
19b												
20	0.11	0.02	0.006	0.81	0.8	0.003	0.03	0.0014	0.014	0.18	19	18
21	0.11	0.01	0.003	0.56	0.8	0.003	0.11	0.0019	0.003	0.33	25	44
22											9	23
23	0.17	0.31	0.008	0.36	1.7	0.008	0.06	0.0114	0.003	0.50	23	21
24	0.03	0.01	0.003	0.19	0.3	0.006	0.22	0.0014	-	0.03	10	9
25	0.03	0.06	0.003	0.19	0.7	0.003	0.19	0.0019	0.007	0.08	8	16

7/3 - 8/3

Station	Br	Mn	V	Al	Fe	Cr	Zn	Sb	Cd	Pb	SM	SO_2
1	0.50	0.18	0.025	1.22	4.1	0.008	1.64	0.0058	0.008	1.25	131	177
2	0.64	0.36	0.078	1.19	2.3	0.019	0.72	0.0044	0.006	1.47	120	195
3											77	144
4	0.53	0.21	0.052	2.03	2.1	0.011	0.58	0.0058	0.006	1.28	101	165
5	0.06	0.34	0.049	0.83	3.9	0.014	2.08	0.0069	0.013	2.33	126	193
6	0.78	0.26	0.043	0.86	2.4	0.008	0.69	0.0069	0.006	2.39	148	200
7											74	133
8	0.44	1.91	0.058	0.83	13.3	0.019	10.22	0.0228	0.028	3.28	133	151
9	0.53	1.26	0.044	0.83	9.7	0.017	12.72	0.0139	0.024	3.61	107	136
10	0.65	0.21	0.042	2.36	2.7	0.009	1.11	0.0088	0.009	1.11	25	108
11											26	77
12	0.22	0.16	0.023	0.58	1.1	0.008	0.44	0.0036	0.003	0.47	42	39
13	0.08	0.16	0.015	0.36	0.6	0.008	0.08	0.0017	0.004	0.23	26	19
14	0.58	0.17	0.033	2.28	1.7	0.022	0.50	0.0108	0.008	1.08	66	83
15	0.25	0.12	0.024	0.44	1.0	0.011	0.39	0.0050	0.011	0.44	62	98
16	0.67	0.02	0.018	0.56	0.3	0.003	0.19	0.0039	0.004	1.69	67	29
17	0.11	0.11	0.013	0.33	1.0	0.006	0.28	0.0025	0.006	1.42	30	37
18	0.19	0.27	0.018	0.50	2.3	0.008	1.33	0.0036	0.010	0.58	30	48
19a												
19b	0.25	0.18	0.021	0.97	2.6	0.006	1.11	0.0025	0.006	0.67	47	70
20	0.50	0.38	0.041	1.94	5.6	0.017	2.44	0.0067	0.008	1.69	89	112
21	0.78	0.31	0.037	1.72	4.3	0.011	1.31	0.0061	0.010	1.67	107	116
22	0.31	0.30	0.039	0.58	2.1	0.011	0.92	0.0031	0.006	1.42	69	157
23	0.78	0.33	0.059	1.94	4.8	0.019	0.42	0.0042	0.004	1.53	114	132
24	0.36	0.23	0.042	0.83	2.7	0.011	0.44	0.0042	0.006	0.72	70	63
25	0.14	0.23	0.031	0.50	1.6	0.006	0.53	0.0025	0.004	0.39	44	40

APPENDIX II

Appendix II. Matrices showing correlation coefficients between various chemical parameters for each individual day and for the composite material. (SM = black smoke)

4/2 - 5/2

SM	1.000											
Br	.933	1.000										
Mn	.637	.482	1.000									
V	.548	.495	.521	1.000								
Al	.683	.749	.079	.398	1.000							
Fe	.659	.560	.955	.629	.235	1.000						
Cr	.671	.580	.955	.586	.180	.974	1.000					
Zn	.561	.439	.973	.500	.060	.956	.950	1.000				
Sb	.745	.753	.456	.395	.714	.537	.506	.438	1.000			
Cd	.630	.556	.724	.297	.276	.720	.717	.741	.613	1.000		
Pb	.959	.944	.708	.631	.669	.768	.772	.673	.755	.683	1.000	
SO_2	.885	.778	.652	.507	.502	.605	.666	.556	.582	.559	.842	1.000
	SM	Br	Mn	V	Al	Fe	Cr	Zn	Sb	Cd	Pb	SO_2

5/2 - 6/2

SM	1.000									
Br	.905	1.000								
Mn	.831	.683	1.000							
V	.908	.848	.868	1.000						
Al	.335	.393	.079	.194	1.000					
Fe	.831	.664	.979	.836	.181	1.000				
Cr	.885	.725	.952	.882	.192	.975	1.000			
Zn	.852	.685	.991	.891	.078	.975	.968	1.000		
Sb	.860	.709	.837	.814	.395	.876	.906	.851	1.000	
SO_2	.912	.891	.729	.864	.393	.758	.814	.741	.766	1.000
	SM	Br	Mn	V	Al	Fe	Cr	Zn	Sb	SO_2

10/2 - 11/2

SM	1.000											
Br	.559	1.000										
Mn	.096	.127	1.000									
V	.207	.337	.589	1.000								
Al	.287	.282	-.154	-.047	1.000							
Fe	.163	.157	.592	.416	.133	1.000						
Cr	.375	.272	.237	.488	.064	.565	1.000					
Zn	-.035	.172	.582	.445	.439	.655	.064	1.000				
Sb	-.046	-.122	-.157	-.060	-.030	.137	.073	-.067	1.000			
Cd	.047	.479	.192	.325	.670	.400	.078	.771	-.032	1.000		
Pb	.205	.636	.035	.268	.695	.230	.003	.593	.004	.900	1.000	
SO_2	.667	.499	.495	.524	.216	.474	.564	.290	.105	.308	.227	1.000
	SM	Br	Mn	V	Al	Fe	Cr	Zn	Sb	Cd	Pb	SO_2

11/2 - 12/2

SM	1.000											
Br	.706	1.000										
Mn	-.206	-.113	1.000									
V	.271	.368	.128	1.000								
Al	.044	.110	-.128	-.133	1.000							
Fe	-.071	.255	.409	.228	.303	1.000						
Cr	.403	.482	.182	.513	.255	.274	1.000					
Zn	-.090	-.027	.250	.098	-.050	.199	.145	1.000				
Sb	-.108	.142	.393	.391	.081	.480	.231	.496	1.000			
Cd	.069	.102	.270	.089	.024	.120	.051	.210	.112	1.000		
Pb	.590	.905	-.003	.297	.195	.299	.593	.027	.169	.116	1.000	
SO_2	.499	.461	-.273	.639	.024	.136	.316	-.137	-.086	-.009	.347	1.000
	SM	Br	Mn	V	Al	Fe	Cr	Zn	Sb	Cd	Pb	SO_2

24/2 - 25/2

SM	1.000											
Br	.956	1.000										
Mn	.294	.359	1.000									
V	.718	.783	.742	1.000								
Al	.464	.435	-.029	.130	1.000							
Fe	.540	.590	.870	.806	.399	1.000						
Cr	.119	.057	.516	.286	.153	.554	1.000					
Zn	.311	.359	.789	.669	.169	.872	.575	1.000				
Sb	.765	.755	.464	.680	.598	.685	.458	.442	1.000			
Cd	-.126	-.132	.256	.226	-.044	.303	.050	.368	-.177	1.000		
Pb	.831	.880	.680	.898	.341	.832	.270	.697	.785	.021	1.000	
SO_2	.838	.707	.265	.633	.357	.447	.078	.223	.658	-.107	.664	1.000
	SM	Br	Mn	V	Al	Fe	Cr	Zn	Sb	Cd	Pb	SO_2

7/3 - 8/3

SM	1.000											
Br	.533	1.000										
Mn	.435	.110	1.000									
V	.671	.491	.439	1.000								
Al	.232	.650	-.040	.451	1.000							
Fe	.542	.227	.944	.471	.149	1.000						
Cr	.486	.353	.508	.719	.504	.551	1.000					
Zn	.371	.089	.919	.288	-.060	.895	.409	1.000				
Sb	.466	.295	.880	.418	.244	.857	.581	.811	1.000			
Cd	.378	.051	.905	.282	-.014	.882	.458	.918	.893	1.000		
Pb	.703	.399	.749	.493	.101	.779	.438	.785	.741	.734	1.000	
SO_2	.849	.434	.319	.742	.340	.411	.485	.269	.380	.317	.586	1.000
	SM	Br	Mn	V	Al	Fe	Cr	Zn	Sb	Cd	Pb	SO_2

Composite material

SM	1.000									
Br	.849	1.000								
Mn	.646	.624	1.000							
V	.550	.431	.415	1.000						
Al	.419	.412	.022	.318	1.000					
Fe	.674	.654	.969	.391	.121	1.000				
Cr	.705	.654	.894	.443	.104	.925	1.000			
Zn	.604	.585	.961	.369	.033	.941	.841	1.000		
Sb	.396	.293	.265	.263	.232	.304	.313	.259	1.000	
SO_2	.814	.678	.511	.672	.420	.530	.561	.479	.273	1.000
	SM	Br	Mn	V	Al	Fe	Cr	Zn	Sb	SO_2