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# SOURCE-RECEPTOR RELATIONSHIPS FOR AIR POLLUTANTS IN THE ARCTIC

by

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ROYAL NORWEGIAN COUNCIL FOR SCIENTIFIC AND INDUSTRIAL RESEARCH

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#### ABSTRACT

Estimated emissions of various trace elements from sources in the U.S.S.R. were used in a single-trajectory model to calculate trace element concentrations at Ny Ålesund, Spitsbergen. The calculated concentrations were compared with the levels measured in March/April 1983. Good agreement was obtained for Sb, As, V, Mn, and Cd.

The origin of arctic aerosols has also been studied with the help of statistical methods, such as principal component analysis, cluster, and time series analyses. The results from these analyses, together with the information on wind trajectories, implicate emission sources in the U.S.S.R. as the most important ones for the arctic air pollution in wintertime.

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### SOURCE-RECEPTOR RELATIONSHIPS FOR AIR POLLUTANTS IN THE ARCTIC

#### **1** INTRODUCTION

An assessment of the contribution of source emissions to the ambient levels of air pollution is a key requirement in arctic air pollution studies. To obtain this information, data from emission inventories of air pollutants may be related to the chemical composition of aerosols measured in remote areas, by applying various long-range transport models. Such studies are of particular importance in the Arctic, due to its remoteness from major air pollution sources and the high stability of arctic atmosphere with its pronounced layer structure, particularly in winter (Pacyna et al., 1985b).

Other studies within the BP Programme (Pacyna et al., 1985b) indicate, that the air pollution observed over Spitsbergen at altitudes below 2.5 km in the wintertime, is almost exclusively due to episodic transport from sources in the northern U.S.S.R. In the present report estimated emissions of various trace elements from these U.S.S.R. sources and a simple trajectory model are used to calculate the trace element concentrations at Ny Ålesund (NYA), Spitsbergen. The calculated concentrations are then compared with measured levels during March/April 1983.

To confirm the origin of the aerosols at NYA in March 1983, the seven-element tracer system by Rahn and Lowenthal (1985), principal component analysis, and other statistical methods have also been used with data from the March/April 1983 measurement campaign, when very high concentrations of several trace elements were observed in the Arctic (Pacyna et al., 1985c).

### 2 TRACE ELEMENT EMISSION IN THE U.S.S.R.

The emission data used were taken from a more detailed report (NILU, 1984), based on production and consumption statistics for the U.S.S.R. (UN, 1981), available information on manufacturing techniques, and the efficiency of emission control installations. In this study the anthropogenic emissions of a number of chemical elements present in fuel, ores and rocks (i.e., As, Cd, Cr, Mn, Ni, Pb, Sb, Se, V and Zn) were estimated for all sources in 12 regions of the U.S.S.R. (see Table 1). The locations of these regions are shown in Figure 1 and Table 2 gives the estimated emissions per unit area, based on the EMEP grid system (150 x 150 km). The emission factors used in the calculations were estimated separately for the different source regions.

The main sources of air pollution in the U.S.S.R. are the coal, oil and gas combustion by electric utilities, industrial processes and traffic. In 1979, more than 80 per cent of the electricity was generated in conventional thermal power plants. Bituminous and sub-bituminous coals were used for 35 per cent, and lignite for 15 per cent of the generation. The remaining 50 per cent of electricity was produced in oil and gas-fired power plants. The emission inventory includes the following industrial processes as well: non-ferrous metal production, pig iron and steel manufacturing, cement and phosphate fertilizer production, wood processing, and pulp chemicals. Refuse incineration was not considered. A major part of the trace element emissions in Table 1 originates from primary non-ferrous metal production. Table 3 gives trace element emissions from individual sources located in the northern U.S.S.R. These are likely the main sources responsible for the high concentrations of air pollutants in the Norwegian Arctic.

#### 2.1 THE KOLA PENINSULA

Two large complexes of copper-nickel metallurgical operations are located in this region. The smelters use ores from the Nikel-Zapoljarnyj and the Monchegorsk mines (CIA, 1978). The annual Ni and Cu production is  $6.5 \times 10^4$  and  $7.5 \times 10^4$  tons, respectively.

Coal and gas are burned in two large power plants in the area of Severodvinsk and Arkhangelsk. Bituminous coal is shipped to these plants mainly from the neighbouring Pechora basin.

Iron ore has been mined at Olengorsk and Kovdor for a long time. The combined output of these mines is estimated to  $1.0 \times 10^7$  tons of concentrate in 1976.

Recently, a large mining complex was inaugurated at Kostomuksha in subarctic Soviet Karelia, with a yearly production of  $3 \times 10^6$  tons of iron pellets during its first phase of development (ANR, 1983a). This will be followed by two other development phases, which are expected to reach a full production capacity of  $9 \times 10^6$  tons. Coking coal for the complex is supplied from the Pechora basin.

The Kola Peninsula contains the world's largest apatite deposits. The current annual capacity of the Apatity mining complex and a mine at Kirovsk is about  $1.5 \times 10^7$  tons of concentrates (CIA, 1978; ANR, 1983a), providing more than 70 per cent of all raw materials for the production of Soviet phosphate fertilizers.

Almost 40 per cent of the total wood pulp production in the U.S.S.R. comes from the Kola Peninsula. A giant pulp and paper facility is located in Svetogorsk, and air pollution problems in this area are enhanced by the combustion of bituminous coal. The production of 1 unit weight of wood pulp and paper requires an estimated average charge of 1 unit weight of bituminous coal. Thus,  $3.5 \times 10^6$  tons of coal are burned annually in the complex.

#### 2.2 THE PECHORA BASIN

The Pechora basin is centred on Vorkuta and Inta. In 1976, 2.6 x  $10^7$  tons of coal were mined in the area, including 1.6 x  $10^7$  tons of coking coal. There are 28 mines in the area (not all are producing) including the Vorgashor mine, the largest shaft mine in the European U.S.S.R. (Meyerhoff, 1983).

The major consumers of coal, mined in the Pechora basin, are the industries of the Kola Peninsula, and the Cherepovets iron and steel plant (Dienes and Shabad, 1979).

In the Pechora basin, the main users of low- and high-grade bituminous hard coal are the thermal power plants in Ukhta and Vorkuta, and the industry (wood processing and pulp chemicals production, mineral and fuel extraction, and construction materials). The production of wood pulp in the Pechora basin is estimated to almost  $2 \times 10^{6}$  tons of pulp a year, requiring about  $2 \times 10^{6}$  tons of bituminous coal annually.

#### 2.3 THE NORILSK AREA

The copper-nickel smelter complex in Norilsk is claimed to be the world's largest copper-nickel smelter, consisting of more than 20 mines, smelters and benefication plants. Yearly capacities are  $5.5 \times 10^5$  tons nickel concentrate, and  $6.5 \times 10^5$  tons copper concentrate (ANR, 1983a, 1983b).

The Norilsk smelter utilizes the Outokumpu flash-smelting method. In this process the roasting and smelting operations are integrated, whereas conventionally they are carried out in separate furnaces (Environment Canada, 1982). Among the advantages of this process, three are particularly important from the air pollutant emission standpoint: low energy consumption, high SO<sub>2</sub> concentration in the flue gas (with a possibility to produce  $H_2SO_4$ ), and high copper concentration in the product. Thus, the emissions of atmospheric pollutants from the Norilsk complex are lower than from other smelters in the U.S.S.R.

Coal from the Tanguska province has been used as fuel for power production at Norilsk. Gradually, it was supplanted by hydroelectric power from the Khantayka River dam, and then by gas from the West Siberian fields (Meyerhoff, 1983). Nowadays, coal is still used as a backup fuel for the nearby Snezhnogorsk hydroplant.

Certain amounts of coal are also burned in a thermal power plant in Norilsk. For the purpose of this report it is assumed that 1 per cent of the total electricity production in the U.S.S.R. is generated in the Norilsk region.

The production of wood pulp in the Norilsk area is at the same level as that in the Pechora basin.

#### 2.4 THE URALS

Substantial amounts of nickel and copper are smelted in the southern and central parts of the Urals. The main copper-nickel mining and smelting complexes are centred in Verchnyy Ufaley, Resh, Khalilovo, Orsk, Sverdlovsk, and Chelyabinsk. The annual production in these complexes is  $6.5 \times 10^4$  tons of nickel and  $3.5 \times 10^4$  tons of copper.

Coal, oil and gas are burned in a number of thermal power plants in the region. Additionally, coal and oil are used in several industrial processes. Several coal districts are located in the south-western part of the West Siberian basin, along the eastern flank of the Urals. The coal is mainly of lignite type, and the oil and gas come largely from the West Siberian basin. The Samotlor oil field in this basin is the largest oil field in the U.S.S.R. (Meyerhoff, 1983). Urengoy in the same basin is the world's largest gas field. It is estimated, that Urengoy soon will supply much of the nations' gas, and significantly contribute to the gas consumed in Europe (Meyerhoff, 1983).

The largest amounts of fossil fuels in the Urals are consumed in the Reftinskaya, Verhnaya, Physchma, Karmanovo, Jushnouralsk, Troizk, and Iriklinskij thermal power stations. All have power outputs larger than 1000 MW. It is assumed, that one third of the total coal and a half of the oil consumption in the U.S.S.R. power plants are used there.

Almost half of the pig iron and steel produced in the U.S.S.R. comes from the factories along the Urals. The main iron foundries and steel mills are located around Serov, Nishniy Tagil, Sverdlovsk, Chelyabinsk, Slatoust, Magnitogorsk, Orsk, Kamensk Uralskiy and Rudnyy. Electric arc and open-hearth furnaces are commonly used in the Soviet steel technology.

## 3 APPLICATION OF EMISSION DATA TO ESTIMATE TRACE ELEMENT CONCENTRATIONS IN THE ARCTIC

Emission estimates can be related either to ambient concentrations measured during selected periods, using wind trajectories (e.g., episodes of long-range transport of air pollutants, indicated by high concentrations of several trace elements), or by statistical analyses of how the concentration of the samples relates to the source emissions.

The origin of the aerosols, measured during episodes of long-range transport of air pollutants, was analyzed with the help of a simple trajectory model (Eliassen and Saltbones, 1982), based on the mass-balance equation:

$$\frac{\mathrm{d}q}{\mathrm{d}t} = (1 - \alpha) \frac{Q}{h} - k \cdot q \tag{1}$$

where:

q = trace element concentration in air, ng m<sup>-3</sup>, t = time, s, Q = trace element emission per unit area and time, ngm<sup>-2</sup>s<sup>-1</sup>, referred to the EMEP grid system (150 x 150 km). h = height of mixing layer, m, k = decay rate for the trace element considered (due to wet and dry deposition), s<sup>-1</sup>, α = fraction of trace element emission deposited in the same grid element as it is emitted. (This local deposition supplements the deposition included in the decay rate, k.)

The mass balance equation is integrated along the calculated trajectories. Since Equation (1) is linear in the concentration q, the contributions from emissions at a given position along a trajectory can be considered separately, and added to give the total concentration. For a trajectory, consisting of N sections or timesteps  $\Delta t$ , one obtains:

$$q(N\Delta t) = q(o) e^{-k} N \Delta t + \sum_{i=0}^{N-1} (1-\alpha) \frac{Q \cdot \Delta t}{h} e^{-k(N-i)} \Delta t$$
(2)

where:

q(N∆t)	Ξ	trace element concentration at the end of the
		trajectory, ng m <sup>-3</sup> ,
q(o)	1	trace element concentration at the start of
		the trajectory, ng $m^{-3}$ ,
Q,	=	trace element emission in the i-th grid square,
T		$ng m^{-2} s^{-1}$ ,
N	11	number of trajectory timesteps,
Δt	=	length of timestep, s.

A part of the emitted trace elements is deposited in the same grid element as it is released. Dry deposition studies in the surroundings of copper and lead smelters (Glowiak et al., 1977; Kubacka et al., 1977; Pacyna et al., 1981) and coal-fired power plants (Pacyna, 1980) in Poland were used to assess this "local deposition" of the pollutants (coefficient " $\alpha$ " in Equations (1) and (2)). In these studies, 11-15% of the trace elements released was found to be deposited in the emission area. This may, however, be an overestimate for emissions from tall stacks, equipped with highefficiency control devices.

As seen from Equations (1) and (2), the model concentrations are inversely proportional to the mixing height "h". Based on an analysis of radiosonde data for Europe by Eliassen and Saltbones, (1982), a constant mixing height of 1000 m was used as a preliminary estimate of "h".

The decay rates "k" of the trace elements for a constant mixing height were based on the dry deposition velocities given in the literature review by Davidson (1980), and by other authors (e.g., Welsh Office, 1975). The following dry deposition velocities for aerosols, containing the respective trace elements, were assumed to be representative for the decay rate calculations in the study region:  $0.1 \text{ cms}^{-1}$  for Cd and Se,  $0.2 \text{ cms}^{-1}$  for As, Pb and Sb,  $0.3 \text{ cms}^{-1}$  for V,  $0.4 \text{ cms}^{-1}$  for Cr and Zn, and  $0.5 \text{ cms}^{-1}$  for Mn and Ni. The presence of pollutants in precipitation is the result of several complex processes, both within and below clouds, and uncertainties still remain as to a detailed description of the scavenging efficiency of precipitation. Our knowledge of these processes is very incomplete, and data of this type are largely absent from the literature. For this reason, wet deposition of trace elements was not considered. In the winter there is very little precipitation in the Arctic, in summer this may possibly result in a slight overestimation of the calculated trace element concentrations.

An other important simplification is the assumption of uniform distribution of the pollutants within the mixing layer. The NILU aircraft measurements in the winter Arctic (Pacyna et al., 1985a) show that the layers of polluted air below 1.5 km altitude can be related to episodes of air mass transport from areas having a potential temperature similar to that in the Arctic, e.g., from the northern U.S.S.R. During episodes the concentrations of anthropogenic pollutants were high throughout this layer, and the chemical composition of the aerosol at 800 m was similar to that measured at ground level. In summer the air next to the ground is generally very clean, except during episodes. It is therefore difficult to asses to what extent the above simplification increases the uncertainties of the model calculations.

The application of emission estimates is complicated by uncertainties in the natural source contributions. The dominant natural source of airborne trace elements in the Arctic seems to be wind-blown dust from the many arid areas in Asia. A long-range transport of Asian desert dust, particularly airborne loess, to the Norwegian sector of the Arctic has been suggested on several occasions (Rahn et al. 1979; Winchester et al., 1984, Raatz and Schnell, 1984: Pacyna and Ottar, 1986), but it is very difficult to assess the amounts of fine, eroded dust that can be transported. Among the trace elements, only the concentrations of Mn and Cr are likely to be enhanced in the arctic air due to this transport (Pacyna et al., 1985c). Winchester et al. (1984) have provided an alternative explanation for the presence of large soil particles in the Arctic: emissions from volcanoes, geysers, or surface/ocean-floor disruptions. Indeed, on February 18, 1983 other satellite observations revealed a large plume of aerosols venting from the flank of the Bennett Island (76.7<sup>0</sup>N, 149.3<sup>0</sup>E). Similar aerosol emissions from the Bennett Island were also observed in early April, 1983. The chemical composition of the arctic aerosol measured at Ny Alesund, Spitsbergen did not, however, show elevated concentrations of Cd, As, Co and Pb which are usually associated with olcanic releases (Pacyna et al., 1985d).

As expected, substantial differences in the Cl concentrations in the arctic summer and winter (Pacyna and Ottar, 1985) show that ice cover is a natural obstacle for emission of seasalt aerosols during winter.

Eight cases in March/April 1983 were selected for the calculations. The cases represent air sampling periods when high elemental concentrations were observed, and the wind trajectories clearly indicated a direct transport from the U.S.S.R. (Pacyna et al., 1985d).

#### 4 COMPARISON OF MEASUREMENTS AND ESTIMATES

Fourteen aerosol samples were collected at the NILU sampling station in NYA during March/April 1983. The high-volume samples were collected on Whatman 41 filters three times a week. Detailed descriptions of sampling procedures and equipment are given in Pacyna et al. (1985d).

The samples were analyzed for 40 elements by instrumental neutron activation analysis (INAA) and particle-induced X-ray emission (PIXE) at the Instituut voor Nucleaire Wetenschappen, Rijksuniversiteit Gent, Belgium. A nitric acid extraction procedure was used for the determination of Cd by atomic absorption spectrophotometry (AAS) at NILU, using a Massman furnace with automatic injection.

The present work focuses on the concentrations of As, Cd, Cr, Mn, Ni, Pb, Sb, Se, V, and Zn. These ten elements originate mainly from anthropogenic sources. Generally, the concentrations of Cr, Ni, and Pb were measured by PIXE, the average of the PIXE and INAA data was used for Zn, and the INAA results were used for As, Cd, Mn, Sb, Se, and V. Details about the experimental INAA and PIXE procedures are given by Schutyser et al. (1978), and Maenhaut et al. (1980; 1981), respectively.

The measured concentrations of the ten elements are shown in Figure 2, together with the estimates from Section 3. The analytical error in the measured concentrations is typically less than 5% for V, 5-10% for the elements As, Mn and Sb, about 10% for Pb, Se and Zn, and 20-25% for Cd, Cr and Ni. It should also be noted that the Cr concentrations for Samples N-6 and N-8 were below the detection limit of the analytical method. In these two cases, the Cr concentrations measured by INAA at the Center for Atmospheric Chemistry Studies, University of Rhode Island, U.S.A., are compared with the estimates.

The most interesting feature of the comparison in Figure 2 is that the estimates are close to the measurements for all metals and samples. The ratios between the measured concentrations of the metals are very similar to the ratios between estimated concentrations. The measured concentrations are higher for Zn and Mn, while the estimated concentrations are higher for Pb, Se, V, Cr and Ni. To quantify the difference between measurements and estimates, the relative difference can be calculated using Equation (3):

$$X = \frac{\frac{C - C}{\text{cal meas}}}{\frac{C}{\text{meas}}} . 100\%$$
(3)

where:

The best agreement between measured and estimated concentrations is obtained for Sb. For this element, the average relative difference amounts to only 14 per cent. This suggests that the estimated emissions for Sb were relatively accurate. The average relative differences for the elements As, V, Mn, and Cd (excluding Sample N-10) are 36, 37, 60, and 75 per cent, respectively. Larger differences are found for a group of elements including Cr, Ni, Pb, Se, and Zn, The possible reasons for these differences include incomplete emission inventory, improperly assumed dry deposition velocities, disregard of wet deposition processes, improperly assumed local deposition values, inaccuracies in the analytical procedures, assumption of constant mixing depth, and inaccuracies in trajectory calculations. Based on the experience gained from studies of long-range transport of trace elements over Europe (Pacyna et al., 1984a), inaccuracies in the emission evaluation and in the dry deposition velocities can be considered to be the main causes for the differences between the measured and calculated values presented here.

To investigate further, whether the differences in Figure 2 are mainly due to incorrect emission inventories, or to inaccurate dry deposition rates, the ratios between measured and calculated atmospheric concentrations for each element were divided by the corresponding ratio for Sb. The results are given in Table 4. A ratio of 1 means that the ratio between measured and calculated concentrations for the element in question is the same as for Sb (ideal agreement). The bottom row in Table 4 gives the average values of the ratios. In calculating these averages, inverse values were used for individual ratios which were lower than 1. As the calculated concentrations are proportional to the emission rates, these averages are a measure for the mean discrepancy (relative to Sb) between the observed atmospheric concentrations and the emission estimates. Assuming the Sb emissions to be approximately correct, a "+" sign before the average indicates an underestimation of the emissions and "-" sign an overestimate relative to Sb. As can be seen from Table 4, the Cr, Ni, Pb, Se, and V emissions seem to be overestimated for the majority of the samples (the Ni emissions in all cases) by a factor ranging from 1.6 for Se to 3.8 for Ni. However, in the case of Sample N-8, the Cr, Pb, and Se emissions are underestimated. The same is also true for the emissions of Pb and V for Sample N-9. It is interesting to note that the air sampled at Ny Alesund (Samples N-8 and N-9) had likely travelled over the Norilsk area. In the other cases, the air masses were passing over emission sources along the Urals, on the Kola Peninsula, and in the Pechora basin. This suggests that the Cr, Pb, Se and V emissions listed in Tables 1-3 are overestimated for the sources in the three latter regions, whereas they are underestimated for the Norilsk area. The Ni emissions are apparently overestimated for all regions. The differences between measured and estimated concentrations of the elements Cr, Ni, Pb, Se and V in Figure

2 show generally the same as the difference between the Sb normalized ratios in Table 4. Incomplete emission surveys rather than inaccurate deposition velocities therefore seem to be the main cause for the discrepancies between the observed and calculated concentrations for these elements.

Also for Zn the ratios in Table 4 are similar to the difference between the measured and calculated concentrations in Figure 2. Thus, the discrepancies should be due to either analytical procedures or an inadequate Zn emission inventory. In a study of the trace element transport over Europe, Pacyna et al. (1984a) obtained a value of 3.3 for the ratio of the measured Zn concentration to the Zn emission, using V as normalizing element, when air masses had passed over the U.S.S.R. and Poland to Birkenes in Southern Norway. It was concluded that the high Zn concentration ratio was either due to contamination of the samples, or that the emissions of Zn had been underestimated. At Ny Alesund two different samplers were used simultaneously, and the In INAA data were compared with AAS results. No significant differences were found, indicating that inadequate emission estimates were responsible for the discrepancies between calculated and measured Zn levels. Two explanations can be offered for the underestimated emissions. First, there may be a natural Zn source, which contributes significantly to the Zn concentration at Ny Alesund. By analyzing size-differentiated aerosols from Ny Alesund, Pacyna et al. (1984b) observed that the Zn size distribution was bimodal. The mode below 0.5 µm EAD was suggested to be the result of anthropogenic sources, and the coarse fraction (>8 µm EAD) was tentatively attributed to natural sources, particularly eroded material from the earth's crust. This hypothesis, however, is not confirmed by the corresponding enrichment factors. The Zn enrichment factors, relative to Fe and average crustal rock (Mason, 1966), range from 104 to 205, and are thus significantly higher than the geometric average of 50, calculated for remote areas by Wiersma and Davidson (1984). High enrichment factors indicate, that anthropogenic Zn sources were affecting air quality during March/April 1983 at Ny Ålesund. Thus, it is very likely that an underestimation of the Zn emission from anthropogenic sources in the U.S.S.R. is the cause of the differences between the measurements and estimates, shown in Figure 2 and Table 4. To ascertain which Zn source from Table 3 may be too low, the Zn data were compared with the As data in Figure 2 and Table 4. Both elements show a somewhat similar behaviour. The As emissions from the anthropogenic

sources on the Kola Peninsula and along the Urals are apparently underestimated, whereas the As release from the Norilsk smelter complex (Samples N-8 and N-9) is slightly overestimated. Table 3 indicates that copper-nickel production is the dominant As source and an important Zn source, on the Kola Peninsula and in the Urals. It may therefore be suggested that the As and Znemissions are underestimated in the two regions.

The measurements and estimates for Cd and Mn (Table 4) do not show any consistent difference one way or another. The difference is rather small, except for Cd in sample N-10. However, the data in Figure 2 seem to indicate that the Mn concentrations obtained from the transport model are underestimated. As there is no consistant difference between the measured Mn concentrations and the Mn emissions (Table 4), it is suggested that the low Mn concentrations of the transport model originate from a too high dry deposition velocity for this element (0.5 cm s<sup>-1</sup>). To see, how much of the discrepancy between the measurements and the estimates can be explained by a different deposition velocity, it was assumed that the dry deposition velocity was no greater than 0.2 cm  $s^{-1}$  for all elements. As a result the calculated concentrations of Mn and Ni have increased by an average factor of 2. The increase in the Cr and Zn estimates was approximately 1.5 times, and in the V concentrations 1.3 times. The dry deposition velocities for Cd, Se, As, Pb and Sb were not changed. By doing this, the agreement between measurements and estimates was significantly improved for Zn and Mn. However, the measurements still remain higher than the estimates, particularly for Zn. This may confirm that the difference between the measurements and the estimates is due to underestimated emissions in the case of Zn, and an improper dry deposition velocity for Mn. Decreased dry deposition velocities for Ni, Cr and V cause larger differences between measured and calculated concentration of these elements, particularly in the case of Ni, indicating that the emissions of this element have been overestimated. The changes for Cr and especially V are far less significant.

The above discussion on source-receptor relationships for trace elements at Ny Alesund has also been presented in an article by Pacyna et al. (1984c).

### 5 TRACER SYSTEMS FOR ASSESSING THE ORIGIN OF ARCTIC AEROSOLS

To trace the origin of remote aerosols, Rahn and Lowenthal (1984) developed a seven-element tracer system, which included Zn, As, Se, In, Sb, non-crustal V and non-crustal Mn. They compared the elemental signature (expressed as element ratios to Se) at the receptor site with those of different source areas. Selenium was chosen as normalizing element, because its concentration tended to be less variable than for other elements. Figure 3 shows the ratios of 6 aerosol elements to Se at Ny Alesund as a function of sample number. It is interesting to note that the ratios to Se tend to be highest for samples having the highest absolute concentrations of the elements (e.g., Samples N-3 to N-5, and N-9 to N-11). Rahn et al. (1983) found that the ratios to Se are significantly enhanced in smelter aerosols. Thus, it can be speculated that the air mass sampled during the periods N-3 to N-5 and N-9 to N-11 had passed over industrial areas with extensive smelter activities.

Average ratios for the elements of the seven-element tracer system and  $SO_4^{-2}$ , Sc and Fe to Se are presented in Table 5 for Ny Alesund and Vardø. The latter station is one of five stations on mainland Norway employed during the BP programme, and may be considered as the primary station for tracing emission sources on the Kola Peninsula. A description of the station is given in Vitols and Wasseng, (1985.) The ratios are compared with data for Barrow, Alaska, and with the arctic smelter signatures (Rahn et al., 1983). The similarity between the mean Ny Alesund and Barrow signatures is particularly striking. This seems to suggest that the trace elements at both sites originated from the same source area. The ratios at Vardø are all a factor of 2 lower than the average Ny Alesund data, except for non-crustal V.

The ratios for Ny Alesund in Table 5 are also in a very good agreement with the tracers for the "Russian" aerosol measured at Birkenes (southern Norway) and Rørvik (southern Sweden) as shown by Pacyna et al. (1984c)

### 6 STATISTICAL ANALYSIS OF DATA FROM NY ÅLESUND AND VARDØ

Statistical methods, such as multiple regression analysis, cluster analysis, time series analysis, principal component or factor analysis, and discriminant analysis have found several applications in studies of the origin of aerosols (e.g., Gaarenstrom et al., 1977; Raabe, 1978; Finzi et al., 1979; Horowitz and Barakat, 1979; Henry and Hidy, 1979; Hopke et al., 1980; Thurston and Spengler, 1981; Heidam, 1982; Lowenthal and Rahn, 1985). In this report, time series, principal component, and cluster analyses were employed to confirm (or reject) the hypothesis, that the Ny Alesund and Vardø were affected in March/April 1983 by the same air masses during episodes of long-range transport of air pollutants from sources in the northern U.S.S.R. to the Norwegian sector of the Arctic.

The concentrations of 29 elements in 14 samples from Ny Ålesund and 13 from Vardø are shown in Figure 4. A 2-sample, two-tailed student's t-test was performed on these concentrations to validate the hypothesis that the concentrations of each element at Vardø do not differ from those at Ny Ålesund. There were only a few significant differences at 5% level for Cd, Na, Mg, I, Cs, Sm, and at the 10% level for Cl. The differences for Na, Mg, I, Cl and Cs can be explained by contribution of seasalt particles to the total amount of particles measured at Vardø. The Sm concentrations are too small to judge their real meaning. The higher Cd concentrations at Vardø indicate an additional emission source of that element around Vardø.

An analysis of linear dependencies between the concentrations of the various elements at Vardø and Ny Ålesund was carried out with the use of correlation coefficients. The correlation coefficients of all elements at Ny Ålesund and Vardø, arranged in a matrix, are presented in Tables 6 and 7, respectively. The analysis of the correlation matrix was performed by principal component analysis. Each diagonal block in Tables 6 and 7 is represented by one principal component. Generally, this component is a linear transform, that translates the original variable to a new variable, in order to maximise its variance. In other words, principal components transform the variables into a new orthogonal system of axes, where the units correspond to the amount of variability. The dimension of this new system is interpreted as a number of factors that have influenced the observations. Examination of the correlation coefficients in Tables 6 and 7 show that the elements can be grouped. The first group includes Al, Mn, K, Sc, Fe, Cs and Sm, which are known to be of soil dust origin. The Cl concentrations at Vard $\phi$  are closely correlated with the concentrations of Na and Mg (correlation coefficients >0.89). The group of anthropogenic elements consists of As, V, Se, In, Sb, Pb, S, Ag, Zn, and partly Mn, K, Cr and Fe.

To identify the various air masses that passed over Ny Alesund and Vardø, principal component analysis (PCA) has been performed on 10 elements: Al, V, Mn, Ni, As, Se, Cd, In, Sb and Pb. The variance matrix with factor coefficients was employed instead of a correlation matrix with values of since the log transform is easier to operate. The factor factors, coefficients (PC) for Ny Ålesund and Vardø are summarized in Table 8. The factors themself represent samples, e.g., F-1 represent samples N-1 and V-1. Factor coefficients (PC) for all elements in 8 samples (from N-2 to N-9 and from V-2 to V-9), were plotted in Figure 5 against the factor coefficients (PC) for Samples N-1 and V-1, respectively. The PC coefficients in Figure 5 form several clusters, each possibly representing a different air mass. It is interesting to note, that clusters for F-3 vs. F-1, and F-5 vs. F-1 plots are smaller than clusters for F-2 vs. F-1, F-4 vs. F-1, F-6 vs. F-1, and F-7 vs. F-1 plots, indicating that air mass observed at Ny Ålesund and Vardø during the N-3, N-5, V-3, and V-5 periods was other than the air masses during other samples. The smallest clusters in F-9 vs. F-1 plots, suggesting that air mass during the N-9 and V-9 sampling periods was also different from air masses during the other samples. It is difficult to judge the origin of the air masses using only data from Figure 5. Since the clusters in plots F-3 vs. F-1, F-5 vs. F-1, and F-9 vs. F-1 are similar, it can only be suggested that the emissions from regions with similar source patterns contributed to the air mass pollution during N-3, N-5, N-8 and V-3, V-5 and V-8 sampling periods. The values of the PC factors in Figure 5 suggest non-ferrous metal production as a main emission source. To be able to say which of the non-ferrous metal production regions should be considered, additional information, e.g., wind tracjectories, is necessary.

Canonical correlations (CC) also measure the dependence of two groups of variables, searching for their similar linear structure. Using data from 10 samples at Ny Ålesund and Vardø, eight of the CC's were 1.00  $\pm$  0.01 and 2

were around 0.001. This indicates that the chemical composition of samples from both stations is very similar, and so must be the emission sources affecting them.

#### 7 CONCLUSIONS

Several methods can be used to assess the origin of aerosols in the Arctic. A simple trajectory model has proved to be very efficient for this purpose. However, the use of the model is limited to days with high concentrations and only slowly changing air trajectories. Emission estimates for certain air pollutants from anthropogenic sources in the U.S.S.R. can be related by means of this model to ambient air concentrations at remote locations in the Norwegian Arctic. The modelled concentrations for March/April 1983 agree well with measured concentrations during episodic long-range transport of air pollutants from northern U.S.S.R. to the Arctic. Thus, the emissions from sources (particularly non-ferrous metal production and fossil fuel combustion) on Kola Peninsula, the Pechora basin, the Norilsk area, and the Urals, are main contributors to the high concentrations of anthropogenic elements at Ny Ålesund, Spitsbergen during the winter half-year.

Emission estimates for Sb seem to be the most accurate, and this element can be used as a reference for calculating elemental ratios. These ratios, often called elemental tracers or indicators, include Zn, As, Se, In, non-crustal Mn, and non-crustal V, related to Sb.

The assignment of the origins of arctic aerosols can be also studied with statistical methods, such as PCA, cluster analysis, and time series analysis. The results from these, together with information on wind trajectories, show that emission sources in the U.S.S.R. are the most important ones for arctic air pollution in wintertime.

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Table 1: Estimated emissions of air pollutants in the U.S.S.R. in 1979  $(t y^{-1})$ .

Region	As	Cd	. Cr	Mn	Nİ	Pb	Sb	Se	>	Zn
The Kola Peninsula	165	29	122	106	645	745	23	26	122	180
The Pechora basin	11.5	3.5	81	42	13	198	12	22	66	56
The Norilsk Area	246	. 26	31	28	935	742	25	33	130	262
The Urals	551	145	1390	1160	1620	9530	96	180	3000	3920
The Yakutsk area	6.5	2	46	42	30	130	+	6	20	26
The Moscow area	16	9	99	56	300	3620	9	22	861	14
The Donetsk area	63	4 0	1050	1100	506	3340	25	112	366	2520
The Kuznetsk area	429	262	456	375	138	3940	24	60	158	8830
The Fergana area	980	274	21	40	210	4540	11	66	766	4550
Caucasus	253	54	Q	30	15	2100	25	32	215	266
The Leningrad area	+	2	17	15	80	1090	2	e	233	20
The Baikal area	55	14	11	39	50	715	11	17	33	88
Total ·	2780	860	3320	3070	4660	30700	350	610	6730	20800

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Region	As	Cd	Сr	Mn	Ni	Рb	Sb	Se	>	Zn
The Kola Peninsula (1)	1.2	0.2	0.9	0.8	4.8	5.5	0.2	0.2	0.9	1.3
The Pechora basin (2)	0.25	0.1	1.8	1.6	1.6	4.4	0.3	0.5	1.5	1.2
The Norilsk area (3)	5.5	0.6	1.0	0.6	21	16.7	0.6	0.7	2.9	5.9
The Urals (4)	3.1	0.8	1.7	6.5	9.0	53.1	0.5	1.0	16.7	21.9
The Yakutsk area . (5)	0.3	0.1	0.9	1.8	1.3	5.6	0.2	0.4	0.9	1.1
The Moscow area (6)	0.2	0.1	0.7	0.6	3.3	39.8	0.1	0.25	9.5	0.8
The Donetsk area (7)	0.6	9.0	9.6	9.8	4.5	29.7	0.5	1.0	8.9	22.4
The Kuznetsk area (8)	3.7	2.3	4.0	3.3	1.2	34.3	0.2	0.5	1.4	77.0
The Fergana area (9)	8.9	2.5	0.2	.0.4	1.9	41.1	0.6	0.8	6.9	41.2
Caucasus (10)	3.7	0.8	0.1	0.5	1.1	30.8	9.0	0.5	3.2	3.9
The Leningrad area (11)	0.2	0.1	0.8	0.7	3.6	49.0	0.1	0.1	10.5	6.0
The Baikal area (12)	2.4	4.0	1.8	1.7	2.2	31.9	0.5	0.75	1.5	3.9

.

Table 3: Estimated emissions of air pollutants in northern U.S.S.R.  $(t y^{-1})$ .

Reg	jion and source	As	Cd	Сr	Mn	Ni	Ρb	Sb	Se	>	Zn
-	The Kola Peninsula										
1.1	Copper-nickel production	154	15	2	2	535	412	14.0	16.0		61
1.2	Fossil fuel combustion	2		34	30	10	24	2.0	4.0		10
1.3	Steel and iron	e		14	37	77	13	2.3	1.5		19
1.4	Phosphate fertilizer	I	10	ł	t	21	e	ı	2.4		80
1.5	Wood processing	9	2	39	37	5	26	4.8	1.7		10
1.6	i Gasoline combustion	1	t	1	I	I	237	1	I		l
	Total	165	29	122	106	645	145	23.1	25.6	122	180
e											
2 4											
2.1	Coal mining and	1			ł		1	I			
	combustion	8.5	2.1	60	22	62	27	2	17.5	6 4	0 4
2.2	Wood processing	e	-	21	19	1	13	4.8	1.1	17	16
2.3	6 Gasoline combustion						158				
	Total	11.5	3.5	81	12	13	198	11.8	22.2	66	56
0											
2	Ine Norllsk area .										
3.1	Copper-nickel production	242	24	e	2.5	900	650	22	24.9		235
3.2	Fossil fuel combustion	-	0.5	89	7.0	11	27	-	e	113	8
3.3	Wood processing	3	1.0	20	18.5	24	13	2.4	1.4	17	19
3.4	Gasoline combustion						53				
	Total	246	25.5	31	28	935	742	25.4	32.6	130	262
. 4	The Urals										
4.1	Copper-nickel production	462	10	5	2	585	1220	41.5	14		444
4.2	Fossil fuel combustion	42	33	150	130	190	173	13.6	57	2730	180
4.3	Steel and iron	14	42	1230	966	240	500	38.5	16	267	3300
4.4	Gasoline combustion						7630				
	Total	551	145	1386	1158	1615	9530	94	180	3000	3920

Sample	As	Cd	Cr	Mn	Ni	Pb	Se	v	Zn
N-3 N-4 N-5 N-6 N-8 N-9 N-10 N-11	2.2 2.4 1.5 2.0 0.7 1.8 1.3 1.2	1.5 2.4 0.8 1.0 0.8 2.0 0.3 0.9	0.3 0.4 0.3 0.3 2.5 0.4 0.3 0.4	0.8 1.2 0.8 1.2 0.9 2.6 0.6 1.2	0.4 0.2 0.3 0.4 0.1 0.2 0.2 0.2 0.3	0.8 0.3 0.6 0.8 1.7 1.8 0.3 0.4	0.5 0.5 1.0 1.1 0.5 0.5 0.5	0.6 0.1 0.5 0.6 1.0 1.7 0.3 0.3	2.0 1.7 1.7 1.8 4.9 5.9 0.9 1.5

Table 4: Comparison of measured concentrations relative to emissions, with Sb as normalizing element.

A "+" before the average indicates underestimated emissions, and a "-" an overestimate. A " $\pm$ " indicates that a given element was overestimated for a part of the samples, and underestimated for the rest of the samples.

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Table 5: Average ratios to Se at Ny Alesund and Vardø, and comparison with data from Rahn et al. (1983)

Arctic smelter 9.7 + 1.4 14.9 + 4.9 2.8 + 0.4 185 + 108 + 18 139 + 20 24 + 6 N = 5 signature 61 1.18 + 0.54 3.8 3.1 5.2 + 2.9 11.9 + 6.9 + 21 (filters) + 20 + 60 30 + 20 N = 215.4 + 6.3 + Barrow 50 46 220 (all samples)  $0.45 \pm 0.24$ 5.9 2.3 + 1.8 3.6 + 1.8 9.8 + 4.4 12.4 + 8.1  $12.8 \pm 5.1$ + 18 50 = 13 Vardø + 6.9 + | 26 96 N (N3-N5 and N9-N12)  $1.10 \pm 0.14$ 20.3 + 4.8 + 1.8  $6.9 \pm 2.5$ 8.9 + 2.1 + 70 + 11 43 + 20 30 + 7 Ny Ålesund 8 IJ 9.6 55 250 N (all samples)  $0.83 \pm 0.36$ 3.5 2.5 Ny Ålesund 2.7 19.3 + 6.3 + 89 + 19 42 + 17 25 + 9N = 145.7 + 1 7.0 + 7.1 + 31 191 so4 (x10<sup>-3</sup>)<sup>b</sup> Element Sc (x10<sup>3</sup>) In (x10<sup>-</sup> , Mn v a x Zn As Fe Sb

 ${}^{a}_{X}$  and  $Mn_{X}$  indicate non-crustal V and non-crustal Mn.

b All sulfur measured in this work was assumed to be present as sulfate.

N = number of samples

H	62.	00	. 4Ŭ	19	.09	.01	-,10	.06	.17	.11	27	62.	.20	- 13	.14	.08	44.	F.	.35	-,01	.26	52	66.	05	22.	13 7 432-	19.	03	1.00
13	10	- 25	23	- 25	23	1.2	68.	21	28	21	.18	.09	- 41	62.	17	. 05	-, 08	1.24	19	.10	42	57.	- 04	.39	10.1	12.	02	- 00	03
E.	. 85	42	123	02.	14.	.43	27	44.	64.	14.	21	.18	46	- 25	.70	.71	06.	60 673	LL.	.10			47) 670	55.	5.	. 74	00.1	- 05	÷ ¢ ;
S	.87	ι co	.99	06.	.83	61.		.76	.89	. 83	- 40	.01	.86	12.	32	. <b>4</b> 5	- 89	.90	50.	22.	. 90	. 33	. 4.3	12.	15.	00.	14	10.	ية. ما: •
<b>p1</b>	.76	3.	- 92	. 89	40.	.86	- 49	11.	.87	500	- 54	00.	26.	. 75	68.	14.	.75	.85	. 38	55.	.86	- 14	.21	19.	00.1	16.	59	10	54
θē	44.	,74	.63	13.	20.	.81	43	. 75	.71	11.	.16	22.	95.	. 16 .	.78	.50	49.	69.	. 63	. 05	. 55	30.	()3	00.	.61	5	62.	100	- 02
1254	12.4	11	1.5	e	.05	01	60	.03	5	. 09	62 -	11.	.20	. 13	. 08	.01	.38	.30	.32	- 90.	.26	10	. 00.1	. 08		24.	113 113	. 50.	5.
88	1.25	.36	32	- 25	- 30	. 29 .	•04	.40	31	.27	. 04	.13	- 15	. 16 -	.20	. 03	. 28	12.	67	+0.	- 32	. 00.	- 10 - 10 - 10 - 10 - 10 - 10 - 10 - 10	. 05	13	.33		61.	10
NZ N	.73	.95	.96	5.	56.	. 98	24	. 83	.97	. 95	12	60.	. 85	117	.84	. 64.	. 73	. 16.	32	.31	. 00.1	.32	. 26 .	. 65	. 85	- 06.	53	-	35.
CO	.31	.12	52.	-29	.02	. 11	. 10	.04	.17	.11.	. 38	. 28	.27	. 19 .	.05	.12	. 18	1-3 C-1	.22	. ŬŪ	E.	- 96 -	.05	. 05	.25	.33	.10	.10 -	.01
끹	.92	5	56.	.90	. 39	. 82	. 43	. 84	.90	.90	.30	.10 -	. 64	- 54	.93	- 66 -	- 26.	16.	.00	.22 1	26.	- 62.	32	- 53 -	.88	.95	11.	- 19 -	
CR	.72	95	.96	197	56.	.85	- 22-	. 88	.96	16.	- 12.	.17	.81	15.	. 84	.40	.74	.00	1 16.	.23	.97	1	.30	. 64	.86	.96	580	104	12.
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NA	10	22.	52.	.30	.17	.17	00	.00	53.	4°) •	.00.	14.	- ch	.76	.02	.22	97.	12.	. 30	- 33	12.	· 04	.29	.16	401- 11-2	10	1.1	. 13	C-1
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(n) SE	60.	.95	.90	\$6.	00.	1 26.		.97	38	16.	- 11 -	.13	18.		39	.50	11.	26-	. 89	20.	26.	- 02.	- 90.	52.	50.	19	14.	- 22.	.09
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12	.82	22.	.00	43°	05.	53	- 02.	28.	94	.90	- 62.		88,	- 65.	50	£5°	.82	69.	.95	29	. 95.	- 22'		1.1	26.	. 99 .	10	5	.40
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Table 6: Correlation coefficients for Ny Alesund.

Table 7: Correlation coefficients for Vardø.

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## Table 8: Factor coefficients (log data) for Ny Ålesund and Vard $\phi$ .

۳.	1.	2	3	4	5	6	7	8	9
				Ny Åles	und				2
AL MNI ASEDN SEDN SEDN SE	$\begin{array}{c} 07973 \\ 07084 \\ 06657 \\ 04503 \\ 06055 \\ 01216 \\ -02081 \\ 00050 \\ 01634 \\ 1.0000 \end{array}$	-1.0000 0.3843 -0.1450 0.1529 0.2736 0.1115 -0.1247 0.0048 0.0795 0.3079	0.0162 0.1865 0.2044 0.0532 -0.0328 0.0097 -1.0000 0.0009 0.0015 -0.4921	-0.1138 1.0000 0.7097 0.9495 -0.5739 0.0492 0.8743 -0.0004 0.0961 -0.9643	-0.2016 -0.1360 0.4813 -0.0047 -1.0000 -0.0589 -0.2039 -0.2039 -0.189 -0.1902 0.6207	0.2386 -0.2257 -0.8097 1.0000 -0.3222 0.4032 -0.2145 0.0143 0.1539 0.1598	-0.3097 -1.0000 0.6251 0.7612 0.4381 -0.7638 -0.0111 0.0032 0.3290 -0.0680	-0.1126 -0.4061 0.4354 -0.0517 0.1378 1.0000 0.0649 0.0140 0.0706 -0.1034	0.0225 0.0598 -0.0230 -0.2275 -0.1954 -0.0161 -0.0097 0.0452 1.0000 0.0247
				Vardø					
AL V MN NI AS SCD IN SB PB	1.0000 0.8260 0.6763 0.7046 0.5539 0.1449 -0.0379 -0.0014 0.1593 0.7162	1.0000 -0.5929 0.0295 0.0495 0.0051 0.1407 0.5691 0.0070 -0.0567 -0.7787	-0.2359 -0.2390 -0.5680 0.5838 1.0000 0.2794 -0.2977 0.0009 -0.0855 -0.2596	-0.0688 0.8809 -0.4125 1.0000 -0.8378 -0.1505 -0.1183 -0.0049 -0.0792 -0.8244	0.5457 -0.4673 -0.2679 0.0895 -0.3098 0.4399 -1.0000 -0.0069 -0.1482 0.2506	$\begin{array}{c} 0.0670 \\ -0.3002 \\ -0.9438 \\ 0.5266 \\ -0.2315 \\ -0.4510 \\ 1.0000 \\ 0.0215 \\ -0.0752 \\ 0.9660 \end{array}$	-0.5268 -0.9301 0.9164 1.0000 -0.3932 0.1890 -0.0032 0.0524 0.4360 0.1279	0.1292 -0.0155 -0.3090 -0.0645 -0.3583 1.0000 -0.1277 -0.0046 0.0929 0.2400	-0.0255 -0.0411 0.2205 0.0972 -0.0494 0.1451 0.0479 0.0267 -1.0000 0.0828



Figure 1: Location of emission regions considered in this report.

- 1. The Kola Peninsula
- 2. The Pechora basin
- 3. The Norilsk area
- 4. The Urals
- 5. The Yakutsk area
- 6. The Moscow area
- 7. The Donetsk area
- 8. The Kuznetsk area
- 9. The Fergana area
- 10. Caucasus
- 11. The Leningrad area
- 12. The Baikal area







Figure 3: Ratios of 6 aerosol components to Se as a function of sample number at Ny Ålesund in March 1983. VX and MnX indicate non-crustal vanadium and non-crustal Mn, respectively.



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Figure 4: Time series of trace element concentrations at Ny Ålesund and Vardø in March 1983.



Figure 4: cont.





Figure 4: cont.



Figure 5: Factor coefficient plots for Ny Alesund and Vardø from trace element concentrations measured in March 1983. (See text for explanation.)



Figure 5: cont.



Figure 5: cont.

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Figure 5: cont.

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RAPPORTTYPE OPPDRAGSRAPPORT	RAPPORTNR. OR 3/86	ISBN-82-72	47-666-5
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\* Kategorier: Åpen - kan bestilles fra NILU A Må bestilles gjennom oppdragsgiver B Kan ikke utleveres C

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