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Assessment of the Impact of Opencast Chrome Mining on the Ambient Air Concentrations of TSP, Cr, Ni and Pb Around A Mining Complex in Northern Finland

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ASSESSMENT OF THE IMPACT OF OPENCAST CHROME MINING ON THE AMBIENT AIR CONCENTRATIONS OF TSP, Cr, Ni AND Pb AROUND A MINING COMPLEX IN NORTHERN FINLAND

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This paper describes the results of an air sampling program designed to evaluate the impact of a chromium opencast mining complex on the ambient air environment at Kemi, Northern Finland. The total suspended particles (TSP) and associated metal (Cr, Ni and Pb) concentrations in the air were determined. The TSP samples were collected simultaneously during one year (2000) at three monitoring stations in the mine area using high volume samplers and glass fibre filters. The heavy metal concentrations were analysed by graphite furnace atomic absorption spectrometry (GFAAS) after dissolution of the filters by an alkali fusion procedure. The impact of the opencast chrome mine on the air environment was assessed by comparing the TSP and Pb monitoring data with the Finnish air quality limit and guideline values. The results showed that the Finnish air quality guideline values, annual mean and the 98th percentile value were exceeded for TSP, but not for Pb. The results also indicated that ambient air TSP and heavy metals concentrations in the air varied considerably within short time intervals (day-to-day fluctuation), as well as by the month, and that the Cr and Ni concentrations in the air correlated strongly with TSP.

Keywords: TSP; Cr; Ni; Pb; Air particulate matter; Chrome mining

INTRODUCTION

Air pollution is an ever-increasing concern. Airborne particles, varying widely in chemical composition, size, shape, homogeneity and concentration, are often referred to as ''dust'', regardless of their origin [1]. Mining operations in general have an adverse impact on the environment. During the individual mining operations, such as blasting, digging, crushing, screening, storing of intermediate and final products, packing, loading, unloading and the movement of vehicles along dusty roads, a substantial

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amount of particulate matter is generated and spread over wide areas [2]. Air pollution monitoring has been carried out extensively in the vicinity of mine complexes in order to clarify either the total suspended particles (TSP) concentrations in the air [3–4] or TSP together with metal concentrations [2,5–7]. According to the environmental impact assessment (EIA) procedure for mining, it is very important to document the normal state of the environment before carrying out an environmental investigation or any expansion in production [8–10].

In Finland, a new Environmental Protection Act has been in force since 1.3.2000. It combines the environmental acts required to meet the requirements of Council Directive 96/61 EC of September 1996 concerning integrated pollution prevention and control [11]. According to the Finnish Environmental Protection Act, the operators of industrial processes and plants are generally obliged by their environmental permits to monitor the processes (operation monitoring), releases (emission monitoring) and impact of their operations on the environment (impact monitoring). The monitoring requirements presupposed by the permit provisions are laid down either in the environmental permits or in separate emission monitoring programmes approved by the competent authority. According to the environmental permits approved by the competent authority of the Lapland Regional Environmental Centre, TSP measurements were part of the impact monitoring program of Outokumpu Chrome Kemi Mine Oy in 2000 (current name AvestaPolarit Oy Kemi Mine).

The heavy metals Cr, Ni and Pb were chosen for elemental analysis, because according to the US Environmental Protection Agency (USEPA) they have a special relevance due to their potential carcinogenic impact [5]. Pb was chosen for elemental analysis, because according to the World Health Organisation (WHO) it is potentially toxic [5], and in addition the European Commission legislation obliges member states to monitor the lead content in atmospheric particulates [12]. In this context it is worth noting, that according to our previous study, chromium is one of the major elements in the ore and therefore its concentration in the air was expected to be relatively high, Ni is deposited extensively in the area around the opencast chromium mine according to woodland moss (Pleurozium screberi) analyses made in 2000 [13]. In addition, in Finland there is the air quality limit and guideline value for Pb, too [19]. This work provided an ideal opportunity to study the impact on the environment of opencast chromium mining prior to the planned expansion of chromite ore production. The study is a part of a major project focusing on the effects of industrial activities (i.e., mines and pulp and paper mills) on the environment in Northern Finland [13–15].

EXPERIMENTAL

Study Area and Pollution Sources

The study area is situated near Kemi (65°44'N, 24°35'E) in the Gulf of Bothnia, northern Finland. The Outokumpu Chrome Kemi Mine Oy (current name AvestaPolarit Chrome Oy Kemi Mine) is a large chromium ore deposit located about 7 km from Kemi. Present ore reserve are 70 million tonnes, and the estimated mineral resources 150 million tonnes. The mine produces approx. 1 million tonnes of chromite ore per year. At the same time, 8 million tonnes of waste rock are removed from the open-cast pits.

Pollutant	Quarry, roads, stores	Crushing	Enrichment plant	Total
Particles (t) SO ₂ (t) NOx (t $NO2$) VOC(t)	45	1.5	0.5	47.9 17.8 ^a $16.6^{\rm a}$ 2.0 ^a

TABLE I Major air pollutants and their sources emitted from the opencast chromium mining complex of Outokumpu Kemi Mine in 2000

and a including emissions from the activities of subcontractors operating in the mine area.

Air pollutants emitted from Outokumpu Chrome Kemi Mine Oy are derived from the various mining operations, such as crushing and enrichment plant, roads, piling stores, quarrying and random sources and emissions from a thermal power stations, as well as from the activities of the subcontractors operating in the mine area. The total emissions into the air in 2000 are given in Table I. The particle emissions from the process (thermal power plant $+$ enrichment plant $+$ cruching) were 2.9 t, which is only 6.0% of the total particle emissions (47.9 t). The main emission sources (45 t) were the quarry, roads and stores, which account for about 94% of the total particle emissions. The annual SO_2 and NO_x emissions have been estimated to be rather similar for many years, but particle emissions from roads are estimated to vary depending on how frequently the roads are watered. There are no other heavy metal emission sources in the vicinity of the mining complex. The nearest industrial emission sources are the pulp and paper mills, Oy Metsä-Botnia Ab Kemi Mills and Stora Enso Oyj Veitsiluoto Mills, situated about 7 km away, but they emit no Cr, Ni or Pb [14].

Sampling Sites

Air sampling was conducted at three sites, MA1, MA2 and MA3. The location of the monitoring stations is shown in Fig. 1. Each station represents a different kind of pollution area. Monitoring station MA1 (Porasydänvarasto) is situated on the northern side of the study area, about 800 m from the opencast mine and about 300 m from a heap of mining spoil. Monitoring station MA2 (Konttori) is situated in the middle of the study area, close to the enrichment plant and the storage site for granulated chromite and close to a local railway line where trains are loaded. This area was estimated in advance to be the most polluted area. Most of the buildings and canteen are also situated in this area. Monitoring station MA3 (Selkeytysallas) is situated in the south part of the study area next to the tailing ponds, about 1500 m from the opencast mine. The dominant wind direction in the study area is south or southwest.

Sampling Procedure for TSP

The TSP samples were collected at each monitoring station using a standard TSP high volume sampler (Wedding $\&$ Associates, Inc.) at a height of 3 m above ground level [18]. The samples were collected simultaneously at each site on Sundays, Tuesdays and Thursdays between 2.1.2000 and 28.12.2000. The sampling time was 24 h. A total of 377 TSP filters were collected. The TSP samples were collected by drawing air through the sampler at a known volumetric flow rate $(104 \text{ m}^3 \text{ h}^{-1})$. The suspended particles were collected on glass fibre filters (Munktell MG 160, 203×254 mm, 75 g m^{-2}).

Abbreviation: $OM =$ Opencast chromium mine, and numbers $1-6 =$ tailing ponds.

FIGURE 1 The map of Finland, and the location of monitoring station MA1, MA2 and MA3 in the Outokumpu Chrome Oy mining complex at Kemi.

Glass fibre filters are widely used for the collection of total suspended particulates with high-volume samplers because they trap particles as small as about 0.3 microns [4,10,16]. Before insertion in the high volume samplers, the glass fibre filters were dried at 110°C for 24 h until a constant weight was reached.

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Analytical Procedure for Cr, Ni and Pb Measurements

The heavy metal concentrations in the TSP filters were analysed by GFAAS (Perkin Elmer 5100 ZL) after decomposition of the 35 mm diameter discs by an alkali fusion procedure [18]. The discs were cut from the TSP filters with a stainless steel circular cutter. First, the discs were digested in a platinum dish with 2 ml of HF (p.a., Merck) and, after evaporation to dryness, the residue was heated for 4 min at 900°C. The residue was then mixed with 1.5 g of sodium carbonate (Suprapur, Merck) and $0.5 g$ of boric acid (p.a., Merck), and fused for 30 min at 950° C over a Bunsen burner. After cooling, the fused sample was dissolved in 3 ml of distilled water (ultrapure) and 3 ml of concentrated $HNO₃$ (Suprapur, Merck). The solution was then diluted to 50 ml with distilled water (ultrapur). The blanks (unexposed filters) were digested simultaneously with the field samples. The analysis was validated by analyzing the reference materials PASC-2 (Marine Sediment) manufactured by National Research Council of Canada and NCS DC 73309 (Stream Sediment) manufactured by China National Analysis Centre (see Table II).

RESULTS AND DISCUSSION

Total Suspended Particle (TSP) Concentration in the Air

Descriptive statistics for ambient TSP concentrations (μ g m⁻³) at monitoring stations MA1, MA2 and MA3 in 2000 are given in Table III. In 2000, the TSP concentrations in the air varied between 2.0–102.0 μ g m⁻³, 2.0–1481.0 μ g m⁻³ and 2.0–72.0 μ g m⁻³ at

TABLE II Means ($n = 2$) and standard deviations (\pm) of Cr, Ni and Pb (mg kg⁻¹) analysis of reference samples PACS-2 (Marine Sediment) and NCS DC 73309 (Stream Sediment) compared with certified values

Element	PACS-2		<i>NDC DC 73309</i>	
	Certified value	Measured	Certified value	Measured
Сr	90.7 ± 4.6	100 ± 2.8	40 ± 1	36.5 ± 2.1
Ni	39.5 ± 2.3	42 ± 5.6	14.3 ± 0.4	18.5 ± 3.5
Pb	183 ± 8	171 ± 2.8	636 ± 10	585 ± 21.2

TABLE III Descriptive statistics for ambient air TSP concentrations $(\mu g m^{-3})$ at monitoring stations MA1, MA2 and MA3 in 2000 (24-h sampling period)

monitoring stations MA1, MA2 and MA3, respectively. The highest daily TSP concentration $(1481 \,\mu g\,\text{m}^{-3})$ occurred at monitoring station MA2 in October. This value is exceptionally high compared to the other data, and was due to exceptional weather conditions when a storm lifted a substantial amount of particles into the air. Thus, our results correspond well with other studies carried out in India at the Jharia Coalfield mining area [2–4] and in Spain at the Donana sulphide ore mining area [5], where the highest TSP concentrations reached a level of $750-1200 \,\mu g\,\text{m}^{-3}$. At the same time, our results for the total mass concentrations of TSP in the air were also relatively high $(102 \,\mu g\,\text{m}^{-3})$ at monitoring station MA1, which is 6.7 times higher than the mean concentration $(15.1 \,\text{µg m}^{-3})$.

According to Finnish Air Pollution legislation [19], the air quality limit value for TSP in the air is 300 μ g m⁻³ (95th percentile, 20°C, 1 atm), and the air quality guideline value is $120 \,\mathrm{\upmu}\mathrm{g}\,\mathrm{m}^{-3}$ (98th percentile, 20 $^{\circ}$ C, 1 atm). In 2000 the 95th percentile values were $39.0 \,\mu\text{g m}^{-3}$, $274.7 \,\mu\text{g m}^{-3}$ and $24.7 \,\mu\text{g m}^{-3}$ for MA1, MA2 and MA3, and the 98th percentile values 76.1 μ g m⁻³, 411.6 μ g m⁻³ and 41.0 μ g m⁻³, respectively (see Table III). Thus in 2000 all the 95th percentile values were below the current Finnish air quality limit value at each monitoring site, but the 98th percentile value at monitoring station MA2 exceeded to Finnish air quality quideline value. Finally, if we compare the annual mean values calculated from the 2000 data for TSP (15.1 μ g m⁻³ for MA1, 80.2 μ g m⁻³ for MA2 and $11.0 \mu\text{g m}^{-3}$ for MA3) with the Finnish annual guideline value of $50 \,\mu g\,\text{m}^{-3}$ (20 \degree C, 1 atm), this guideline value was exceeded in 2000 only at monitoring site MA2.

Seasonal variation can be easily estimated by computing monthly average values from the individual 24h data. The monthly average results in 2000 for TSP (μ g m⁻³) in the air at monitoring stations MA1, MA2 and MA3 (Fig. 2) show that the TSO concentrations varied considerably from month to month, especially at monitoring station MA2. If we compare the TSP annual mean $(80.2 \,\mu g \,\text{m}^{-3})$ at monitoring station MA2 to the individual monthly average concentrations, the months when the average means were over 10% higher than the annual mean were May, June, July, September and October. The values during these months were 27%, 28%, 68%, 20% and 159% higher than the annual mean respectively. At monitoring station MA1, the monthly

FIGURE 2 Monthly (January–December) average ambient air TSP concentration (μ gm⁻³) at monitoring stations MA1, MA2 and MA3 in 2000.

FIGURE 3 Day-to-day fluctuation in ambient air TSP concentrations (μ gm⁻³) at monitoring station MA2 between 3. and 28.9.2000.

average means were correspondingly $> 10\%$ higher than the annual mean (51.1 μ g m⁻³) in May, June and October, which were 69%, 31% and 45% higher than the annual mean value. At monitoring station MA3, the monthly average means $> 10\%$ higher than the annual mean $(11.0 \,\mu g\,\text{m}^{-3})$ were recorded in May and October. The results shown in Fig. 2 evidence, that the ambient TSP concentrations were the highest during May–June in 2000 at all the monitoring stations, as well as in July at MA2. The most reasonable explanation for the high summertime values is the dust and particulate material emissions from the surrounding land, piling stores and during loading; in summertime the land, piling stores and ore are not frozen as in wintertime.

In addition to high seasonal variation, large day-to-day fluctuations were also in evidence. Figure 3 shows an example of how TSP in the air at monitoring station MA2 varied during September in 2000. In September, ambient daily TSP concentrations varied between $8-186 \,\mathrm{\mu g\,m}^{-3}$, and the monthly average value was $96.6 \,\mathrm{\mu g\,m}^{-3}$. Thus the highest daily TSP concentration $(186 \,\mu\text{g m}^{-3})$ in September was 92.5% higher than the monthly average $(97 \,\mu g\,\text{m}^{-3})$, and 23.2 times higher than the lowest daily concentration value $(8 \mu g m^{-3})$.

The ambient air TSP concentration at different monitoring stations were strongly dependent on the wind direction (Fig. 4). Thus, daily TSP concentrations which were higher than the annual means occurred at monitoring station MA1 when the wind direction was between $115-270^{\circ}$, between $30-240^{\circ}$ at monitoring station MA2, and between $0-45^{\circ}$ and $100-330^{\circ}$ at monitoring station MA3. These results indicate that the dust emissions at monitoring station MA1 are most likely emitted from a heap of mining spoil located at a distance of 300 m from the station. The most likely dust source for TSP at monitoring station MA2 are the mineral piles at the enrichment plant and activities at the railway line where trains are loaded. In this connection it is worth noting that the highest daily TSP concentration (1481 μ g m⁻³) at monitoring station MA2 occurred under a dominant wind direction of 136°, which indicated emissions from the mineral piles at the enrichment plant.

Ambient Air Cr, Ni and Pb Concentrations

The concentrations of Cr, Ni and Pb were analysed only at monitoring station MA2 because, according to the TSP measurements this was the most polluted area and

FIGURE 4 The correlation between TSP (μ g m⁻³) and wind direction at monitoring stations MA1, MA2 and MA3 in 2000.

most of the anthropogenic activities were situated in the vicinity of this station. The levels of heavy metals were analysed each month from the TSP filter with the highest TSP concentration, apart from September and October when the TSP filter with the second highest value was analysed. Figure 5 shows the seasonal variation in ambient air Cr concentrations (μ g m⁻³) at monitoring station MA2 between January–December in 2000. The ambient air concentrations of heavy metals varied considerably from month to month. In January the lowest monthly value in 2000 was the Cr concentration of $0.65 \,\mu g \,\text{m}^{-3}$ and the highest the Cr concentration of $48.2 \,\mu g \,\text{m}^{-3}$ in July. The average Cr concentration was $17.9 \,\mu g \,\text{m}^{-3}$. Figure 5 also shows that the Cr concentrations were the highest between May–August, as well as in November. The Ni concentrations in the air during January–December in 2000 varied between $0.05-14 \,\mu g \,\text{m}^{-3}$ (average 0.52), and the Pb concentrations were all under 0.1 μ g m⁻³.

If we compare the average Cr and Ni concentrations in ambient air (i.e., $17.9 \,\mu g m^{-3}$) for Cr 0.52 μ g m⁻³ for Ni) to the forthcoming EU directive on PM 10 (20 μ g m⁻³ for Cr and $0.02 \mu g m^{-3}$ for Ni) our results are very high. In this context it is worth noting that PM 10 is only a fraction of TSP. However, atmospheric particulate range $\leq 10 \,\mu m$ (PM 10) is inhaled into deeper respiratory tract resulting in pathologies associated with aerosol pollution [5]. For this reason, the US Environmental Protection Agency (USEPA) promulgated in 1984 an air quality standard for environmental particulate matter based on the measurements of PM 10 instead of total suspended particles (TSP). The EU has also developed a new directive for the monitoring of PM 10 instead of TSP which will be applied in 2005 [20]. However, in Finland there are no air quality limit values for Cr and Ni at all yet [19].

Querol *et al.* [5] has also reported high ambient air heavy metal concentration values for As, Pb and Cu especially during summertime (i.e., July–August). This phenomenon for our results, may be partly due to fact that in summertime the land, piling stores and ore are not frozen, and according to Infante *et al.* [21] partly also in terms of climate, highly influenced by sea breeze, in which the air particulates are thought to be rapidly mixed by wind action. The Finnish Meteorological Institute has also reported this phenomenon occurring especially at summertime at the coastline on the Gulf of Bothnia on studies of aerial distribution pattern of malodourous sulphur compounds emitted from pulp and paper mills located at Kemi [22].

FIGURE 5 Ambient air concentration of Cr $(\mu g m^{-3})$ at monitoring station MA2 during January– December in 2000.

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In this context it is worth noting, that the heavy metals concentrations in ambient air are different at various mine areas depending on the type of ore, operations, processes, weather conditions, the location of sampler, etc, so the results of various studies are not necessarily comparable to each other. Ghose *et al.* [23] has reported lead concentrations between 0.4–6.6 μ g m⁻³, and Cr concentrations between 0.004–0.08 μ g m⁻³ in ambient air at coal mine operations in India. Correspondingly very high metal concentrations in ambient air in mine areas can also occur. Querol *et al.* [5] has reported exceptionally high maximum metal concentrations in Donana area (sulphide ore) at Spain: $2.7 \mu g m^{-3}$ for As, and $4.4 \mu g m^{-3}$ for Pb. However, the maximum Cr concentration in that study was $0.07 \,\mu g \,\text{m}^{-3}$, and the maximum Ni concentration $0.04 \,\mu g \,\text{m}^{-3}$.

The cross-correlation plots between the TSP and Cr, Ni and Pb ambient air concentrations shown in Fig. 6 clearly indicate, that the Cr and Ni concentrations in the air increased strongly with the TSP concentration ($R = 0.842$ and $R = 0.862$, respectively), but not for Pb $(R = 0.394)$. The high Kendall correlation between Cr and Ni concentration in the air $(R = 0.758)$ indicates that these metals originated from the same emission sources in the area [24]. However, the correlations between Cr and Pb $(R = 0.238)$, and between Ni and Pb $(R = 0.333)$ were lower.

According to Fig. 6 it is also possible, that Pb in total suspended particles in originated at least from two different emission sources, i.e., emissions from the quarrying and other mine operations, and emission from dust derived from road traffic, but these are impossible to distinguish from each other at this context. However, it is worth noting, that the exhaust emissions from road traffic does not consist of lead (Pb), because the petrol used by automobiles and vehicles in Finland is unleaded [25].

In Finland there are no air quality limit values for Cr and Ni, but there is for Pb $(0.5 \,\mu\text{g m}^{-3})$ and this was not exceeded in 2000. Because the highest Pb concenctration in the air in mine area reached a maximum value of $0.09 \,\mu\text{g m}^{-3}$, which is only 18% of the air quality limit value of $0.5 \mu g m^{-3}$, and simultaneously the correlation between Pb and TSP was rather poor $(R = 0.394)$, we conclude that the air quality limit value for Pb is not likely to be exceeded in the mine area at the current level of operations.

Validity of the Alkali Fusion Procedure

The recoveries of Cr, Ni and Pb after the alkali fusion procedure from the reference materials Marine Sediment PASC-2 and Stream Sediment NCS DC 73309 are given in Table II. The alkali fusion procedure gave a higher result for Ni in NDC DC 73309 than the certified value. In addition, the alkali fusion procedure gave lower results for Pb especially in NDC DC 73309. However, the values for Cr, Ni and Pb after the alkali fusion procedure were all statistically acceptable for reference material. In this context it is worth noting, that Yamashige *et al.* [18] have reported the volatilization of Pb at high temperatures during the alkali fusion procedure. This phenomenon has also been reported in other corresponding studies [26–28]. However, in this context the alkali fusion procedure had to be used, because according to Yamashige *et al.* [18], and Wang et al. [16,29] low recoveries for Cr are expected by Acid decomposition methods. In addition, the use of alkali fusion procedure is accepted by the competent authority of the Lapland Regional Environmental Centre. However, if the volatilization of Pb has happened for real samples (i.e., for total suspended particulate matter collected at mine area), then the calculated ambient air concentrations for Pb are higher that we have expressed.

FIGURE 6 Cross-correlation plots between the TSP and Cr, Ni and Pb concentrations in ambient air at monitoring station MA2 in 2000 ($n = 12$).

CONCLUSIONS

The study shows that mining operations have an adverse impact on the environment. The impact of opencast chromium mining by Outokumpu Chrome Mine Oy on the air environment is clearly visible in the area surrounding the mining complex. In 2000, the opencast chromium mine had the greatest influence on the air environment at monitoring site MA2 (Konttori), especially during May–July and September– October, when the monthly average means were correspondingly 27–67% and 17–158% higher than the annual mean $(80.2 \,\mu\text{g m}^{-3})$ at this site. In 2000, both the annual mean $(50 \,\mu g\,\text{m}^{-3}, 20^{\circ}\text{C}, 1 \text{ atm})$ and 98th percentile value $(300 \,\mu g\,\text{m}^{-3}, 20^{\circ}\text{C},$ 1 atm), which are air quality guideline values for TSP in Finland, were both exceeded at monitoring site MA2. During 2000 the Finnish air quality guideline value for Pb $(0.5 \,\mu\text{g m}^{-3})$ was not exceeded. Our results, for the Cr and Ni concentrations in ambient air were higher but Pb concentrations were lower compared with other studies at mine areas.

Because TSP and heavy metal concentrations in the air can vary considerably over short time intervals (day-to-day fluctuation), as well as monthly, TSP measurement should be carried out as a long-term study in order to obtain reliable data about TSP concentrations in the air. It is also worth nothing that chromite ore production is relatively constant throughout the year, and difference in production could not explain therefore the difference in TSP and heavy metal concentrations in the air between summer and winter. Finally, according to the Pb concentrations measured in the mine area, there is no likelihood that the current level of operations will exceed the level of $0.5 \mu g m^{-3}$, which is the air quality limit value set on the basis of health criteria.

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