

Assessment of historical exposures in a nickel refinery in Norway

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Objectives The aim of the study was, on the basis of new information on nickel species and exposure levels, to generate a specific exposure matrix for epidemiologic analyses in a cohort of Norwegian nickel-refinery workers with a known excess of respiratory cancer.

Methods A department-time-exposure matrix was constructed with average exposure to total nickel estimated as the arithmetic mean of personal measurements for periods between 1973 and 1994. From 1972 back to the start of production in 1910, exposure concentrations were estimated through retrograde calculation with multiplication factors developed on the basis of reported changes in the metallurgical process and work environment. The relative distribution of water-soluble nickel salts (sulfates and chlorides), metallic nickel, and particulates with limited solubility (sulfides and oxides) was mainly derived from speciation analyses conducted in the 1990s.

Results The average concentration of nickel in the breathing zone was ≤ 0.7 mg/m³ for all workers after 1978. Exposure levels for smelter and roaster day workers were 2—6 mg/m³ before 1970, while workers in nickel electrolysis and electrolyte purification were exposed to concentrations in the range of 0.15—1.2 mg/m³. The level of water-soluble nickel was of the same order for workers in the smelting and roasting departments as in some of the electrolyte purification departments.

Conclusions Compared with earlier estimates, the present matrix probably offers a more reliable description of past exposures at the plant.

Key terms exposure matrix, monitoring, multiplication factors, nickel industry, occupational exposure, personal measurements, retrospective exposure assessment, speciation.

Since the 1930s an elevated risk of respiratory cancer has been demonstrated among nickel-refinery workers in several countries (1—5). The epidemiologic analyses, however, were restricted to variables such as worksite, job type, date or duration of employment, and other time variables. No epidemiologic analysis with quantitative data on nickel exposure existed until specific exposure characterizations were introduced in a large reassessment organized by the International Committee on Nickel Carcinogenesis in Man in 1990. The Committee compiled and reevaluated updated results from 10 cohorts of nickel-exposed workers to obtain more information on the carcinogenic effect of different forms of nickel (6).

In refineries nickel is found in a variety of compounds, by convention classified according to their chemical properties. The International Committee on Nickel Carcinogenesis in Man reassessed the cancer hazard according to the following 4 categories of nickel:

water-soluble [the most common forms being nickel chloride (NiCl₂) and nickel sulfate (NiSO₄)], sulfidic [typically subsulfide (Ni₃S₂)], metallic, and oxidic nickel [eg, nickel oxide (NiO)]. The Committee concluded that exposure to oxidic, soluble, or to the combination of sulfidic and oxidic nickel was likely to cause respiratory cancer.

It has been claimed that the epidemiologic findings were somewhat impaired by more recent studies on rats and mice experimentally exposed to aerosols discontinuously for 2 years. These studies supported earlier experimental findings that nickel subsulfide had the most pronounced carcinogenic effect, while no effect was found from the inhalation of nickel sulfate (7). The apparent contrast to the conclusions from the International Committee on Nickel Carcinogenesis in Man could be due to deficiencies in the assessment of exposures among nickel workers, or it could be perceived as a display of the limitations inherent in animal experiments.

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The results from the Norwegian cohort of nickel-refinery workers contributed strongly to the observed association between cancer and exposure to water-soluble nickel. This hazard has been ascribed to the high exposure levels in the electrolysis departments of the Norwegian nickel works in comparison with the levels found at refineries with similar technology. Still, only dust and total nickel had been determined in aerosols of the Norwegian plant before the study by the International Committee on Nickel Carcinogenesis in Man, and the exposure assessment relied mainly on a qualitative approach. Estimates of the 4 types of nickel compounds according to time period and department (or group of departments) were agreed upon by a panel of experts who chose between 5 given ranges of concentration. These categories were subsequently transformed into mean values taken to be the average of the end points of the ranges, rendering the levels 0, 0.3, 1.3, 5, and 10 mg/m³ (6). During the last decade, however, several samples have been analyzed with respect to specific forms of nickel. The results have revealed that the composition of aerosols can differ markedly from what was believed in earlier years.

An elevated risk of respiratory cancer has been found for Norwegian workers with their 1st employment in the nickel industry even in the period 1968 to 1978 (8). Thus, in contrast to many other historical high-risk operations, a period bearing clear indications of carcinogenic exposure now partly overlaps the time span covered by personal measurements, which have been regularly conducted at the refinery since 1973. These circumstances call for a better nickel exposure assessment and a renewed epidemiologic follow-up. The aim of our study was to develop an exposure matrix based on the quantitative use of personal monitoring, direct analyses of individual nickel species in dust and aerosols, and historical reports on the production and work environment. The matrix and subsequent epidemiologic studies were expected to contribute to the discussion of what concentrations and types of nickel exposure represent a cancer hazard.

Material and methods

Production methods

Since the start in 1910 the Norwegian refinery has based its production on the treatment of a sulfidic nickel copper concentrate (matte). Until 1978 the process followed the Hybinette method, including grinding, conversion of the metal sulfides into oxides through roasting, leaching (ie, extraction) of copper from the metal oxides with dilute sulfuric acid, and subsequent smelting of impure nickel anodes from the leach residue (undissolved material). Copper was produced directly by electrolysis (electrowinning) of the filtrate (solution) from the leaching

step. Nickel was deposited in a very clean form on cathodes inside of filtercloth bags (Hybinette bags), which were constantly filled with purified nickel electrolyte. Outside these bags, but still inside the same tanks, the impure anodes dissolved, yielding nickel ions and small amounts of other metals to the solution. This electrorefining process thus required a steady draining of impure electrolyte from the nickel tanks and a continuously operated purification circuit, with precipitation and filtration steps, until pure nickel electrolyte could be recycled to the Hybinette bags (9).

Regarding occupational exposures, the most essential change in the original production process was the abandonment of the Hybinette method in 1978, along with its highly contaminated roasting, smelting, and copper cementation steps. Since that year the refinery has been running a chlorine leach process based on the extraction of nickel directly from ground matte by means of chlorine (10, 11). However, the chlorine leach residue, still containing some nickel, has been subjected to roasting, and copper has been extracted from the roasted leach residue with sulfuric acid and electrolyzed, according to the same principles as in the old Hybinette era. Another important change in the process was the introduction of chlorine gas in 1952. This step allowed for cobalt production, and, as a desirable side effect, the problem with the build-up of arsenic was solved. The notable changes in technology include the replacement of a manually rabbled horizontal roaster furnace with a mechanically rabbled multiple hearth oven in 1915, the introduction of modern fluid bed roasters along with the chlorine leach process in 1978, and a method for copper cementation (precipitation) which was employed in the electrolyte purification circuit between 1930 and 1978, based on the production of finely divided metallic nickel to be added to the impure electrolyte.

The water-soluble nickel compounds comprised predominantly sulfates until 1978, except for some smaller facilities like the cobalt-extraction and chlorine-regaining section from 1952 on. Since 1978 soluble nickel has been found as sulfates in the roasting, smelting, precious metal, copper leaching, and copper electrolysis departments, while the nickel electrolysis and closely associated departments have been dominated by nickel chloride. For our study, the 2 species are referred to as soluble nickel.

Assessment of exposures

This paper covers nickel and nickel compounds, and no attention has been paid to any other potentially harmful exposure. The material for the investigation included more than 500 stationary dust or nickel determinations from the years before 1973, some 5900 personal measurements of total nickel in the breathing zone from 1973 through 1994, and a few analyses of nickel species in dust

and aerosols in the 1990s. In addition, some central documents prepared for the International Committee on Nickel Carcinogenesis in Man (6), along with some new historical reports, were made accessible for our study.

Stationary samples do not necessarily give a representative picture of the nickel inhaled by a worker who is moving around between areas and job tasks with ever-changing aerosol concentrations. Personal sampling, on the other hand, constitutes a better approach for the valid quantification of inhalation exposure. Personal monitoring at the present refinery represents a uniform and long-term documentation of exposures. Therefore, we chose to start the exposure assessment with the measurements from the most recent period. The entire procedure comprised the following 5 steps: (i) the calculation of average levels of total nickel in departments monitored with personal measurements between 1973 and 1994, (ii) a widening of the matrix to include departments with no available measurements, (iii) retrograde extension of the total nickel levels to periods before 1973, (iv) development of speciation keys (ie, the relative amounts of water-soluble, sulfidic, metallic, and oxidic nickel), and (v) a final calculation of exposure levels based on the estimates of total nickel and the proportions of nickel species according to department and time period.

Personal monitoring, 1973–1994. The earliest personal measurements were part of a 1973 investigation by the National Institute of Occupational Health, a well-designed survey with 400 personal measurements conducted together with 200 8-hour stationary samples from the same days and departments as the personal ones. The results were reported directly to the refinery and were made available for our study. The rest of the measurements originated from routine personal samples from most of the contaminated departments, repeated nearly once a year. All available personal measurements were performed with a 37-mm filter cassette, and the nickel concentrations were determined by atomic absorption. The recordings included date, start and stop time of pump (alternatively duration of pump activity), department name, job task, day or shift work, and nickel concentration in milligrams per cubic meter. All the samples were taken outside respirators or masks, even in high exposure situations where the use of respirators would be compulsory.

The measurements were grouped by department according to the historical personnel files. Compared with earlier exposure estimates, some further resolution was introduced through a larger number of single departments and differentiation between shift and day workers for some operations. The personal measurements had an average pump running time of 390 minutes (6.5 hours), and they were all transformed into 8-hour time-weighted

averages, presuming zero exposure when the pump was not running. The measurements in each department were displayed in box-and-whisker plots according to calendar year between 1973 and 1994. Trends in the median values were evaluated by inspection of the graphs and time shifts introduced whenever there was an apparent persistent change in the exposure level. Departments with 10 or less measurements were excluded from further analysis. In a few departments, there were single years in which the median deviated markedly. These measurements were included in the final calculations only when experienced company staff would judge them to be representative.

On the basis of information about work conditions, we presumed that most of the variance within each department and time period was due to day-to-day variation. Thus the arithmetic means of the values should constitute relevant exposure estimates for the calculation of cumulative exposures in epidemiologic studies (12). A large set of records such as ours was expected to comprise a number of erroneous or highly unrepresentative recordings in which spills or outbursts had contaminated the filter during sampling. It was not possible to trace these incidents individually, and, in order to attenuate their potentially strong influence on the arithmetic averages, we trimmed the data through the exclusion of nickel values above the chosen limit of 50 mg/m³ (nickel). In the most contaminated areas this concentration would correspond to a total dust level of 150–200 mg/m³ (dust), which would hardly be tolerated throughout a workday. This step led to a loss of only 6 measurements (0.1% of all) in the range 53–277 mg/m³ (nickel).

There were 220 measurements (3.7% of all) noted to be below the detection limit, and these recordings were substituted with a value that was half the detection limit. We explored the effect on the mean exposures when either the value 0 or the limit of detection itself was used as a substitute and found the difference to be negligible.

Departments with no measurements, 1973–1994. Production or service departments with no available measurements were assigned an exposure identical to departments with a similar work environment. Alternatively, if the environment was better described as an intermediate between 2 departments, the average of the corresponding exposures was taken as the best surrogate. Maintenance workers were, as a rule, assigned the exposure experienced by production workers in the departments they served. Some groups of skilled workers and shops had limited though recurrent contact with the process or production equipment. Depending on the frequency and type of contamination, they were judged to have exposure levels in the range of 0.01–0.05 mg/m³. Clerical workers and administrative personnel were considered to be unexposed.

Retrograde calculation of the total nickel levels. All exposures to total nickel before 1973 were estimated through retrograde calculation with multiplication factors. The principal source of information for the estimation of these factors was the preparatory documents developed for the International Committee on Nickel Carcinogenesis in Man (6). Important changes in production technology and chemistry, as well as ventilation and other environmental improvements, had been described by experienced engineers, and the corresponding influence on exposures had been discussed and summarized by an expert panel (6, 8). In our study, however, it was possible to use a larger number of time shifts and to treat departments separately rather than in groups. In addition, in 6 of the most heavily contaminated departments, stationary measurements from 1969 to 1972 were available in a number of 37 to 166 recordings per department. The recordings were mainly 24-hour samples collected during repeated surveys in the same exposure areas. The average levels were compared with the stationary measurements from the comprehensive 1973 investigation and used as indicators of relative changes in exposure. When no shift in exposure could be inferred from available data, we chose to retain the estimates from the more recent period. A draft of the matrix was evaluated by an advisory group with knowledge of the occupational hygiene, epidemiology, and local production history.

Relative amounts of nickel compounds. Recent speciation analyses have demonstrated the presence of water-soluble nickel in dust and aerosols from roasting and smelting operations that have previously been regarded as practically devoid of these compounds (13, 14). The composition of the samples was determined through a sequential leaching procedure according to Zatka et al (15), or, alternatively, a simplified version of the method (13). Our estimates of the distribution between the 4 groups of nickel compounds were based primarily on speciation analyses from the Norwegian refinery (table 1)

and some measurements from a Russian nickel works on the Kola Peninsula (14). In some departments with process steps unique to the old Hybinette era, speciation analyses of aerosols had never been performed, and in these instances we had to construct ratios based on the matrix in the report from the International Committee on Nickel Carcinogenesis in Man (6) with slight adjustments according to our present knowledge that nickel is usually found as a mixture of all 4 kinds of compounds.

Results

The calculation of the average exposure levels from personal measurements supplied the matrix with data for 29 of a total of 86 departments, including the main production areas known to have had the heaviest exposures and the largest number of employees. Results from the most important departments are shown in table 2. In 16 departments, mainly service units, the exposure levels were estimated based on similarity with respect to work environments. Another 22 low-exposure areas were assigned values between 0.01 and 0.05 mg/m³. Finally, in 19 departments, the workers were considered to be unexposed. The highest average nickel levels according to personal measurements were found in departments with roasting, calcining, and smelting processes. The maximum concentration in these areas was about twice as high as the highest value found in other departments from the same period. The matte crushing and grinding department was the most contaminated department in the 1980s and later, with a nickel concentration of 0.7 mg/m³.

Exposure concentrations before 1973 were derived through back calculation with multiplication factors. Special attention was paid to the early 1970s when the results from the 1st Norwegian cancer study was published. In 3 of the 6 departments with more than 30 stationary measurements taken between 1969 and 1972, the

Table 1. Proportions of water-soluble nickel salts in dust samples from the roasting and smelting departments of a Norwegian nickel refinery. (NIPERA = Nickel Producers Environmental Research Association; IOM = Institute of Occupational Medicine, Edinburgh, United Kingdom)

Sampling method	Site, source or situation	Soluble nickel (percent by weight)	Reference
Stationary samples	Cleaning of roaster, 3rd floor	30	Internal report, 1989
Old dust deposits	Smelting department	20	Internal report, 1991
Personal samples	Demolition of old smelter	4	Internal report, 1991
Dust from electrofilters	Exhaust from roasters	40 ^a	Internal report, 1994
Personal samples	Roasting and smelting departments	10	Internal report, 1995
Stationary samples	Roasting department, 3rd floor	43	Internal report, 1996
Stationary samples	Roasting and smelting departments	4.5–35	Andersen et al, 1998 (13)
Personal samples	Roasting and smelting departments	26 in inhalable fraction, 45 in thoracic fraction, 72 in respirable fraction	Aitken et al, 1998 (report to NIPERA from IOM)
Stationary samples	Smelting department	7–9	Internal report, 1998

^a Water-soluble metal salts, most of it probably nickel sulfate.

Table 2. Arithmetic mean (AM) and geometric mean (GM) of personal measurements of total nickel in air (mg/m^3) by department and time period with the corresponding geometric standard deviation (GSD) and number of samples included (N). The results were based on measurements from a survey made by the National Institute of Occupational Health (1973) and from the refinery's own routine recordings (1973–1994).

Department	Period	AM	GM	GSD	N
Crushing, grinding	1973–1992	0.73	0.32	3.76	296
Calcining, smelting (shift)	1973–1977	0.59	0.27	3.73	136
	1979–1994	0.40	0.13	4.33	133
Calcining, smelting (day)	1973–1977	1.51	0.64	3.78	96
	1979–1994	0.53	0.20	3.62	296
Roasting (shift)	1973–1977	0.58	0.42	2.14	93
	1979–1994	0.19	0.07	4.19	75
Roasting (day)	1973–1977	1.90	1.12	2.75	84
	1979–1994	0.38	0.10	4.60	296
Old smelter building (no 1)	1973–1977	2.56	0.91	3.62	18
Mechanics	1975–1989	0.39	0.10	4.20	65
Copper leaching	1973–1977	0.87	0.32	3.74	113
	1979–1988	0.35	0.17	3.11	146
	1989–1994	0.11	0.05	2.59	149
Copper electrolysis	1973–1986	0.13	0.05	2.68	135
	1987–1990	0.03	0.03	1.51	107
	1991–1994	0.02	0.01	1.67	123
Copper cementation	1974–1977	0.58	0.27	2.99	101
Cobalt separation	1974–1977	0.32	0.12	3.36	36
	1979–1984	0.20	0.07	2.47	97
Cobalt refinery	1983–1994	0.01	0.01	2.31	316
Nickel electrolysis	1973–1977	0.12	0.08	2.16	147
	1978–1980	0.11	0.08	2.58	60
	1983–1986	0.05	0.03	2.28	243
	1987–1994	0.03	0.02	2.14	354
Chlorine leach	1976–1994	0.11	0.03	2.97	585
Electrolyte purification	1974–1977	0.18	0.16	1.69	90
	1979–1986	0.15	0.07	2.98	228
	1987–1989	0.05	0.03	2.21	94
	1990–1993	0.03	0.02	2.02	82
Tankhouse basement	1973–1989	1.15	0.12	4.71	48
Matte leach	1973–1994	0.17	0.04	3.66	519
Filtercloth shop	1975–1993	0.05	0.03	2.49	48
Shearhouse	1973–1993	0.06	0.02	2.79	227

average of the measurements was more than 2.5 times higher than the corresponding results from the 1973 survey made by the National Institute of Occupational Health. Based on an informal comparison of the means, supplemented by reports on technical changes and production problems, the exposure levels during the former period were considered to be 1.5 and 2 times higher in the roasting and smelting departments and the copper cementation area, respectively.

In another area, the matte leach plant, a reduction by a factor of 2 in the average nickel level was observed when 54 personal measurements from the 1973 survey by the National Institute of Occupational Health were compared with the recordings made during subsequent years. The decrease was interpreted as a downward trend in exposure, and a factor of 2 was estimated for the back calculation of concentration levels.

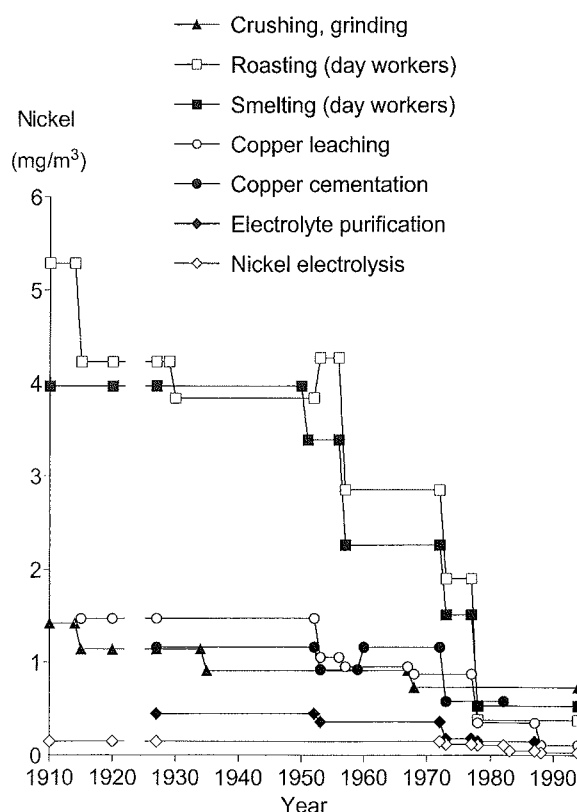


Figure 1. Time trends for the concentration of total nickel in air in selected departments.

After the process change in 1952, the nickel content of the copper electrolyte was reduced from around 70 g/l to approximately 40 g/l. Since aerosols from the electrolyte constituted the main air contamination in the copper tankhouse, a factor of $70/40=1.75$ was used for calculating nickel exposures prior to 1952. The estimation of the rest of the multiplication factors was based mainly on descriptions of changes in process technology. Knowing that the most radical change took place in 1978, and because of uncertainty, we made conservative estimates that resulted in factors within the range of 0.75–1.5. At the most, 5 such multiplication factors were introduced for a single department during the period 1910 to 1972.

During the total time span from 1910 to 1994, the highest exposures were found in the smelting and roasting departments in the 1950s and earlier, with concentrations between 2 and 6 mg/m^3 (figure 1). Among the hydrometallurgical processes, the highest exposures to nickel occurred in the copper leaching and the copper cementation departments, with values around 1 mg/m^3 in the 1960s and earlier. All departments were assigned zero exposure between 1921 and 1926 when the refinery was shut down due to economic problems. During the Second World War, the production rate was lowered, and more than half of the employees became redundant. The technical methods, however, were the same, and there

was no indication of improvement in the work environment. Thus the exposure levels for active workers during the war were considered to be the same as in the periods before and after the war. Due to the general uncertainty regarding exposures before 1973 and the use of carefully prepared historical reports, there was no major disagreement in the advisory group when a draft of the matrix was discussed, and only minor adjustments were necessary.

For the main production departments the relative amounts of different nickel compounds in aerosols were estimated on the basis of determinations from recent years. Up to 3 different historical speciation keys were developed for some departments due to technical or chemical changes. The ratios of nickel species in selected departments are shown in table 3. The proportion of water-soluble nickel was estimated to be 10–15% of the total nickel values in the crushing, roasting, and smelting departments.

Discussion

Exposure assessment is a central component in most etiologic epidemiologic research, and the concentration levels and the quality of the data are important elements in risk assessment. In our study exposures to total nickel back to 1973 were based on a large number of personal measurements of aerosols, which is a preferred sampling method for surveying inhalation exposure. Before arithmetic means were calculated, the data were trimmed gently through the exclusion of 6 obviously erroneous or highly unrepresentative measurements. The high variability observed in some departments was not surprising, due to the different exposure situations that have occurred during normal operation, production problems, and maintenance work.

The assessment of exposures through back calculation with multiplication factors was used for the period 1910–1972. This method has been discussed and applied in earlier works (16, 17), and problems with uncertainty, potential misclassification, and scarce possibilities for validation have been underlined. The risk of misclassification is impending when several multipliers are introduced for the same subgroup. In our matrix, no more than 5 multipliers of moderate size were introduced for a single department. This practice led to a maximum 3-fold relative change compared with the earliest period with personal measurements. In contrast, based on the personal measurements from 1973 to 1994, the average nickel concentration was reduced by a factor of 5, 6, and 8 in the roasting, electrolyte purification, and copper leaching departments, respectively. This decrease reflects, above all, the great production changes in 1978, which affected several of the most-contaminated departments.

Table 3. Distribution of the different forms of nickel in air, estimated as the proportion (by weight) of total nickel, by selected departments and time periods. The estimates were based on speciation analyses in the 1990s, measurements from a Russian nickel refinery (14), and exposure estimates from a study made by the International Committee on Nickel Carcinogenesis in Man (6).

Department	Period	Form of nickel			
		Soluble	Sulfidic	Metallic	Oxidic
Crushing, grinding	1910–1994	0.12	0.72	0.11	0.04
Old smelter building (no 1)	1910–1929	0.10	0.05	0.01	0.84
	1930–1950	0.10	0.05	0.08	0.77
	1951–1977	0.10	0.04	0.18	0.68
Calcining, smelting	1951–1977	0.10	0.05	0.01	0.84
	1978–1994	0.12	0.13	0.01	0.74
Roasting	1910–1977	0.10	0.15	0.03	0.72
	1978–1994	0.15	0.05	0.00	0.80
Copper leaching	1910–1994	0.49	0.01	0.01	0.49
Copper electrolysis	1910–1994	0.80	0.04	0.04	0.13
Copper cementation	1927–1977	0.45	0.05	0.45	0.05
Electrolyte purification	1927–1977	0.80	0.03	0.15	0.02
	1978–1994	0.98	0.01	0.00	0.01
Nickel electrolysis	1910–1977	0.87	0.05	0.01	0.08
	1978–1994	0.83	0.04	0.02	0.11

Forty-three stationary measurements from an early survey by the National Institute of Occupational Health in 1964 (18) suggested that exposure levels in the early 1960s were about the same as around 1970. The absence of any major change would be in accordance with our estimates.

A few reports from other nickel refineries contain exposure data that can be compared with our matrix. In 1984, Warner (19) reported estimates of airborne nickel based on gravimetric personal measurements that were not far from the values found for the 1960s and 1970s with our matrix, except for the higher levels in the Norwegian nickel anode smelting department (table 4). Exposures in nonsintering departments at a refinery in Port Colborne (Ontario, Canada) were presented by the International Committee on Nickel Carcinogenesis in Man (6). Their estimates may also have been of the same magnitude as the Norwegian ones, although only a rough comparison was warranted (table 4). Still, the Norwegian copper leaching and copper cementation departments showed considerably higher levels than those reported for general electrolysis in Port Colborne. Kiilunen and her co-workers (20) published results from personal monitoring in the electrolysis department (nickel electrowinning) of a Finnish refinery. The higher averages in the Finnish plant around 1980 can partly be explained by the generation of aerosols from tanks with no surface additives. Oil or polystyrene balls were applied to reduce this problem in the Norwegian plant as early as the 1950s. Thomassen and his co-workers (14) recently reported nickel exposures in a Russian refinery running a Hybnette process. The Russian roasting and smelting

Table 4. Concentration of total nickel in air in different refineries by department, presented as the arithmetic mean of personal measurements or the range of arithmetic means. Levels before 1973 in the present refinery were estimated through backwards calculation from more recent periods.

Refinery	Nickel in air	Results from corresponding departments in the present study	
		Nickel in air (mg/m ³)	Period
Warner, 1984 ^a (19)			
Grinding	0.10—0.47	0.73—0.91	1951—1977
Electrolyte purification	0.14—0.24	0.18—1.2	1951—1977
Nickel tankhouse	0.05—0.19	0.12—0.15	1951—1977
Shearhouse	0.03—0.14	0.06	1951—1977
Casting of nickel sulfide ingots	0.26—0.40	0.40—0.53	1979—1994
Nickel anode smelting	0.34	0.59—3.4	1951—1977
Port Colborne, 1960s and 1970s (6)			
Electrolysis (general)	<0.25	0.12—0.28 ^b	1951—1977
Nickel anode smelting	<5.0	0.59—3.4	1951—1977
Outokumpu, 1979—1981 (20)			
Nickel tankhouse	0.16—0.23	0.12—0.28 ^b	1951—1977
Monchegorsk, 1990s (14)			
Roasting	6.3	0.58—4.3	1951—1977
Nickel anode smelting	5.1	0.59—3.4	1951—1977
Nickel tankhouse	0.11	0.12—0.15	1951—1977
Electrolyte purification	0.13	0.18—1.2	1951—1977

^a Refineries not indicated.

^b Range including measurements from copper electrowinning and nickel electrorefining.

departments showed values approximately 1.5 times as high as the Norwegian ones in the early 1950s, and the highest level of water-soluble nickel in the Russian refinery was reported for the roasting department (14). In our study as well, the level of water-soluble nickel in the roasting and smelting departments equaled the concentrations in some highly contaminated hydrometallurgical departments. Aitken reported, in 1996, an inverse relation between the amount of soluble nickel and particle size in samples from the Norwegian plant (report to the Nickel Producers Environmental Research Association from the Institute of Occupational Medicine in Edinburgh, United Kingdom); this finding agrees with earlier statements that fine particles are sulfated in the flue from roasters and smelters (19).

Among the personal measurements, we would expect the presence of a higher number of unrepresentative values than the 6 extremes that were excluded from the data before the analysis. The effect of the exclusion of the single highest measurements within each department was explored, and in 3 cases such exclusion would have lowered the arithmetic mean by 50% to 60%. Most of the means, however, would have remained unchanged or would have been reduced by less than 20%.

For some departments there were large discrepancies in the absolute concentrations of nickel in air when our

matrix was compared with the corresponding exposure estimates referred to in the report by the International Committee on Nickel Carcinogenesis in Man (6). Nevertheless, when time trends of the midrange values in the report were considered, the relative change backwards from 1973 was more or less of the same order, although there were differences in the timing of the shifts. In the 1996 update on cancer incidence in the Norwegian cohort (8) the exposure assessment mainly relied on the matrix from the International Committee on Nickel Carcinogenesis in Man, but some of the low-exposure departments were assigned a lower value. Our matrix confirmed the earlier suspicion that some of the estimates have been too high. The lower levels make the cancer hazard in the Norwegian cohort a matter of more universal interest, as the concentrations are closer to those reported from other kinds of metal work, for instance, in the plating industry and welding (21, 22).

Several aspects of our assessment strategy suggest that our matrix represents an improvement. Basically, we calculated exposure levels from a large number of personal measurements, which covered the periods before and after the most radical process change in 1978, and thereby linked the whole matrix to a quantitative analysis. Second, the introduction of a higher degree of resolution with respect to time periods and departments was expected to reduce misclassification further. Third, the proportions of different types of nickel species relied on direct analyses of dust and aerosol samples and thereby reduced the degree of subjective judgment. We therefore conclude that the new matrix probably offers a more reliable description of past exposures at the refinery.

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