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RIJKSINSTITUUT VOOR VOLKSGEZONDHEID EN MILIEU
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RIVM report 729999 002

Assessment of air quality for arsenic, cadmium, mercury and nickel in the Netherlands

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Abstract

The presence of arsenic, cadmium, mercury and nickel in air in the Netherlands has been investigated. Using measurement data, a limited supplemental monitoring effort and the results of modelling calculations, it has been possible to obtain a realistic picture of air quality in the Netherlands with respect to these substances. Background concentrations of arsenic, cadmium and nickel as derived from measurement results range from 0.6-1.1 ng m^{-3}, 0.2-0.4 ng m^{-3} and 2.0-2.5 ng m^{-3}, respectively. Air quality figures for mercury as derived from model calculations are 2-3 ng m^{-3}. Increased cadmium concentration levels in industrial areas were observed near the Hoogovens (Steelworks) and in the Rijnmond industrial area. Nickel levels increased in the Rijnmond area and were shown to be in good agreement with model calculations. All levels were usually found to be within the air quality objectives. Exceptions here are cadmium and nickel, for which the Negligible Risk levels are locally exceeded in comparing these levels to air quality objectives.

Samenvatting

De ondersteuning van de discussie in Europees verband over de invulling van de dochterrichtlijnen van de Europese Kaderrichtlijn Luchtkwaliteit vereist o.a. informatie over de luchtkwaliteit voor arseen, cadmium, kwik en nikkel in Nederland. In dit rapport worden de resultaten van het onderzoek dat is uitgevoerd naar de luchtkwaliteit van deze stoffen, besproken. Een belangrijk aspect van het onderzoek is om potentiële probleemsituaties te inventariseren.
Bij het onderzoek is primair gebruik gemaakt van bestaande informatie om de luchtkwaliteit te karakteriseren, Het onderzoek is aangevuld met een beperkte meetinspanning voor die situaties waar op grond van vooronderzoek is gebleken dat de aanwezige kennis te gering is. Deze meetinspanning was gericht op het verbeteren van de kennis over de situatie nabij een aantal brongebieden.
De luchtkwaliteit kan getoetst worden aan een aantal luchtkwaliteitdoelstellingen.

Arseen concentraties in lucht in Nederland worden vooral bepaald door de grootschalige achtergrond, die in de orde van 0,6-1,1 ng m^{-3} ligt. Er is een gradiënt over Nederland van zuid ('hoog') naar noord ('laag'). In industriegebieden treden nauwelijks verhogingen op.
Voor *cadmium* bedraagt het grootschalig achtergrondniveau in Nederland 0,2 tot 0,4 ng m^{-3}. Ook hier wordt dezelfde gradiënt als voor arseen vastgesteld. Een lokale verhogingen in de orde 100% is vastgesteld nabij de Hoogovens. Een lichte verhoging wordt waargenomen in het Rijnmondgebied.
Het achtergrondniveau voor nikkel is in Nederland 2,0-2,5 ng m^{-3}. In het Rijnmondgebied blijken de nikkelconcentraties verhoogd te zijn: 5-15 ng m^{-3}. Deze resultaten komen goed overeen met de resultaten van de modelberekeningen.
Uitspraken over de luchtkwaliteit voor *kwik* zijn geheel ontleend aan de uitkomsten van modelberekeningen. Het grootschalige achtergrondniveau bedraagt ca. 2-3 ng m^{-3}. Lokale verhogingen zijn op grond van beschikbare emissiegegevens niet te verwachten.
Vergelijking met luchtkwaliteitdoelstellingen leert dat de in Nederland voorkomende niveaus veelal onder luchtkwaliteitdoelstellingen liggen. Uitzonderingen zijn cadmium en nikkel waarvoor lokaal de niveaus van het Verwaarloosbaar Risico worden overschreden.

De natte arseen- en cadmiumdeposities laten geen ruimtelijke afhankelijkheid over Nederland; ze liggen in de orde van 60-240 resp. 80-240 µg m^{-2} j^{-1}. De natte depositie van kwik bedraagt ca. 20 µg m^{-2} j^{-1}. De natte depositie van nikkel bedraagt ca. 640 µg m^{-2} j^{-1}. In het Rijnmondgebied en onmiddellijke omgeving worden verhoogde natte nikkel deposities waargenomen tot ca. 1000 µg m^{-2} j^{-1}.

Summary

To support the discussion in the European context on the daughter directives of the European Framework Directive on Air Quality will require such data as information on the air quality related to such substances as arsenic, cadmium, mercury and nickel in the Netherlands. This report will discuss the results of the investigation carried out on the quality of the environment when affected by these substances. It is important to inventory the results for potential problem situations.

The investigation made primary use of available information to characterise air quality. The research was supplemented with limited monitoring efforts for the situations in which pre-studies had shown the present knowledge to be insufficient. This monitoring effort was focused on improving the knowledge about a situation near a number of source areas. Air quality can be tested for several air quality objectives.

Arsenic concentrations in air in the Netherlands are mostly the result of their large-scale background levels, ranging from 0.6- 1.1 ng m^{-3} for a gradient across the Netherlands reaching from south ('high') to north ('low'). Increased concentration levels in industrial areas were practically non-existent. The large-scale background levels for *cadmium* came to 0.2 to 0.4 ng m^{-3}, where the same gradient as for arsenic was set. An increased local cadmium level in the order of 100% was found near the Hoogovens (Steelworks) and a slight increase observed in the Rijnmond area. The background level for *nickel* is 2.0-2.5 ng m^{-3}, with increases of 5-15 ng m^{-3} nickel observed in Rijnmond. These results are in good agreement with model calculations. Air quality figures for *mercury* originate exclusively from model calculations, with large-scale background levels representing approximately 2-3 ng m^{-3}. Based on emission data, local increases are not anticipated. These levels are usually found to be within the air quality objectives. Exceptions here are cadmium and nickel, for which the Negligible Risk levels are locally exceeded in comparing these levels to air quality objectives.

The wet depositions of arsenic and cadmium across the Netherlands do not allow any spatial dependency; these deposition levels are in the order of 60-240 μg m^{-2} j^{-1} and 80-240 μg m^{-2} j^{-1}, respectively. The wet deposition of mercury comes to ca. 20 μg m^{-2} j^{-1}. In the Rijnmond area and immediate surroundings, nickel depositions of up to 1000 μg m^{-1} j^{-1} have been observed

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1 Introduction

The ‘Framework Directive on ambient air quality assessment and management’ came into force in 1996 (EU, 1996). Consequential to this general European guideline, a number of daughter directives for specific components would come into force in the course of the next few years. For example, daughter directives have recently been implemented for the substances in the so-called first phase*:* sulphur dioxide, fine particulate and lead (EU, 1999). Proposals of the European Commission for the substances in the second phase, ozone, benzene and carbon monoxide, are under parliamentary discussion. Position papers are in preparation for the remaining substances, or third phase, i.e. polycyclic aromatic hydrocarbons (PAHs), cadmium, arsenic, nickel and mercury, to serve as the basis for EC proposals for the daughter directives. This last group may be facilitated through opinion-forming if, for example, sufficient factual information on the presence of these substances in air is available. To meet the last objective cited, the National Institute of Public Health and the Environment (RIVM) carried out a project in 1998/1999 to establish rulings on air quality for PAH, arsenic, cadmium, mercury and nickel; for PAH the focus will be on benzo[a]pyrene. An important aspect will be to inventory situations where raised levels of substances might occur. This report (729999 002) will deal with arsenic, cadmium, mercury and nickel, while in a separate report (729999 001) PAHs are discussed.

Chapter 2 of this report will discuss the general set-up, and the methods and materials used in the research, followed by a discussion in Chapter 3 of the results of model calculations and monitoring. Chapter 4 contains the discussion of these results and Chapter 5 the conclusion to the report findings. The more general environmental aspects of arsenic, cadmium, mercury and nickel will not be discussed in this report; please refer to more general publications on arsenic (Slooff *et al*., 1990), cadmium (Ros *et al*., 1988), mercury (Slooff *et al*., 1995) and nickel (Slooff *et al*., 1992).

2 Material and methods

2.1 Research outline

The starting point for this assessment was that use should be made of information already available, especially that derived from measurements. This could possibly be supplemented with the results of model calculations. Dispersion models were supposed to be used to detect potential problem situations near industrial sources. After this, the remaining incompleteness in the information could be examined with a limited monitoring programme. The project plan was formulated and carried out according to this outline. The model instruments will be explained in §2.2, and operational and recent measurement activities will be presented in §2.3. This information has resulted in the definition of limited RIVM monitoring programme, which will be explained at the end of §2.3.

2.2 Model calculations

The model calculations, with which concentration levels due to industrial sources were estimated, were carried out using the Operational Priority Substances model. Van Jaarsveld (1990; 1995) has described this model. Input data on emission strength, chimney height and heat content for the model were taken from the Dutch Emission Registration. Further details can be found in Appendix B.

2.3 Measurements

The inventory of operational and recently performed monitoring activities includes:

- Measurements of the provincial authority of Limburg for one urban location. These are 24-hour samples, taken every three days.
- Measurements of the provincial authority of Noord-Holland near the Hoogovens (Steelworks). These are 24-hour samples, taken every eight days.
- Measurements by the provincial authority of Zuid-Holland at a number of regional locations and one industrial location. These are 24-hour samples, taken every six days.
- Measurements by RIVM through the Dutch National Air Quality Monitoring Network, taken at three regional and one urban location. These are 24-hour samples, taken every two days.

Measurements of these metals found in precipitation are carried out within the Dutch Air Quality Monitoring Network, as well as in some of the regional networks.

The results to be reported in this publication came into being in different ways. For this reason measurement results are not directly comparable. Appendix C gives details on sampling and analysis methods.

The inventory of monitoring activities showed only a limited number of measurements in industrial environments. Furthermore, the model calculations (see further in § 3.1) showed enhanced levels for nickel, and to a lesser extent for cadmium; in two industrial areas. These observations led to the defining of supplemental cadmium and nickel measurements by RIVM on two industrial sites: Beverwijk, near the Hoogovens (Steelworks), and Schiedam in the Rijnmond area. The usual sampling period of 24 hours was chosen. Sampling was done with a PM10 inlet according to the current views on exposure. A characterisation of measurements and monitoring sites of all the institutions/networks is given in Table 1. Figure 1 shows the locations of the sites.

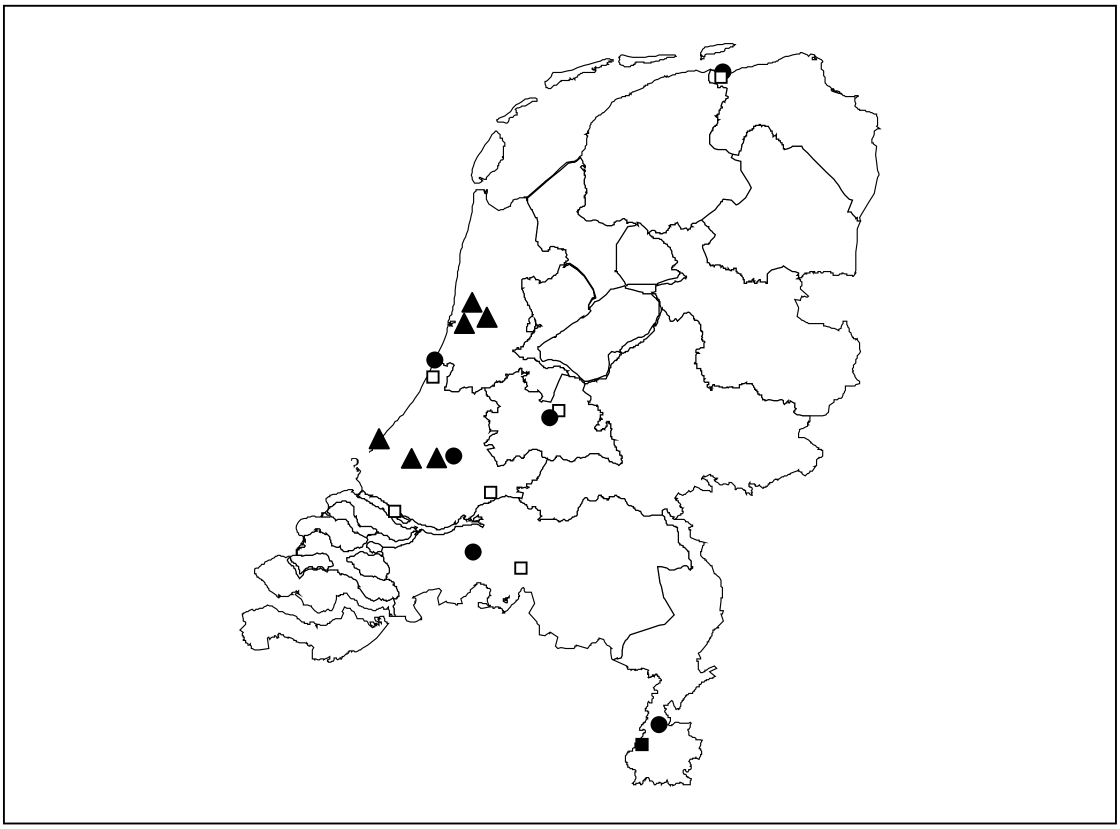

Figure 1 Locations showing operational or recent measurements of arsenic, cadmium, mercury and/or nickel [☐ regional background ▲ industrial area ■ urban background ● wet deposition].

Table 1 Measurements of arsenic, cadmium, mercury and nickel reported on here[1].

Location[1]	Components	Type[2]	Matrix[3]	Time period
Province of Limburg				
Maastricht	As, Cd, Ni	Urban background	A	1/1996-12/1997
Province of Noord-Holland				
IJmuiden	As, Cd, Ni	Industrial area	A	1/1998-12/1998
Wijk aan Zee	As, Cd, Ni	Industrial area	A	1/1998-12/1998
Province of Zuid-Holland				
Bergambacht	As, Cd, Ni	Regional background	A	1/1996-12/1998
Hillegom	As, Cd, Ni	Regional background	A	1/1996-12/1998
Hoek van Holland	As, Cd, Ni	Industrial area	A	1/1998-12/1998
Korendijkse Slikken	As, Cd, Ni	Regional background	A	1/1995-12/1998
Westland	As, Cd, Ni	Regional background	A	1/1996-12/1997
National Institute of Public Health and the Environment (RIVM)				
Beek (133)	As, Cd	Regional background	P	As, Cd: 1/1993-12/1998
Beverwijk (555)	As, Cd, Ni	Industrial area	A	9/1998-6/1999
Biest-Houtakker (230)	As, Cd, Ni	Regional background	A	As, Cd: 1/1987-12/1998 Ni: 1/1998-5/1999
Bilthoven (627)	As, Cd	Regional background	A	As, Cd: 1/1987-12/1998 Ni : 1/1998- 5/1999
De Bilt (628)	As, Cd	Regional background	P	As, Cd: 1/1993-12/1998
De Zilk (444)	As, Cd	Regional background	P P	As: 1/1994-12/1997 Cd: 1/1995-12/1998 Hg: 1/1995-12/1998
Gilze-Rijen (231)[1]	As, Cd	Regional background	P	As, Cd: 1/1993-12/1998
Kollumerwaard (934)	As, Cd, Ni As, Cd	Regional background	A A P	As, Cd: 1/1987-12/1998 Ni: 1/1998-5/1999 Cd: 1/1993-12/1998
Rotterdam (434)	As, Cd, Ni	Industrial area	P	As, Ni: 1/1993-12/1997 Cd: 1/1993-12/1998
Schiedam (439)	As, Cd, Ni	Industrial area	A	9/1998-6/1999
Vlaardingen (433)	As, Cd	Urban background/ Industrial area	A	As, Cd: 1/1987-12/1998 Ni: 1 /1998-5/1999

[1] Not all measurement activities of the networks are shown in this table. RIVM and the provinces of Limburg and Zuid-Holland measure arsenic and cadmium in precipitation at other locations.

[2] Type according to network.

[3] A: air, P: precipitation.

3 Results

3.1 Model calculations

3.1.1 Large-scale background

Results of investigations on the large-scale dispersion pattern of *cadmium* and *nicke*l are described in Baart *et al*. (1995). The model calculations were, however, carried out with emission data from 1990. For this reason source receptor matrices from this investigation were used to make an update, in other words, the most recent emission data are used to calculate new concentrations for the Netherlands**[1]**. This has shown that because of the substantial emission reductions in the neighbouring countries (with respect to the Netherlands), the concentrations for cadmium and nickel, and in the Netherlands for cadmium as well, has been considerably reduced, leading in 1990 to a large-scale background level of 0.6 ng m^{-3} for cadmium and 4.6 ng m^{-3} for nickel. In 1995 these levels had dropped to 0.3 ng m^{-3} and 3.6 ng m^{-3}, respectively. The contribution of Dutch sources to the average concentration level in the Netherlands is estimated at 10% for cadmium and 60% for nickel.

Results of model calculations for *mercury* at European scale were published by Petersen *et al*. (1995) and Ryaboshapko *et al*. (1998), and for the NW European scale by Baart *et al*. (1995). The European background concentration of ca. 1 ng m^{-3} mercury in air and 3-5 ng l^{-1} in rainwater can be derived from these studies. The concentrations calculated for the Netherlands come to ca. 2-3 ng m^{-3} and 20-50 ng l^{-1}, respectively. Mercury, which is supposed to be present as metallic mercury in the atmosphere, is characterised by a relatively long atmospheric residence time and large-scale dispersion pattern. Therefore the calculations also show most of the deposition of mercury in the Netherlands to be caused by foreign sources (see Table 2).

Table 2 Origin of the total mercury deposition (in %) in the Netherlands (Ryaboshapko et al., 1998).

Country of origin	Contribution
Germany	35
The Netherlands 1)	17
Belgium	13
France	6
Great Britain	6
Other anthropogenic sources	8
Natural sources	15

1) The total deposition for the whole of the Netherlands is 2338 kg.

No results are available from large-scale dispersion calculations for *arsenic*.

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[1] Emission reductions for *cadmium*: Belgium (1990-1997) 77%, Germany (1990-1995) 65%. France (1990-1995) 65%, The Netherlands (1990-1995) 30%
Emission reductions for *nicke*l: Belgium (1990-1997) 68%, Germany (1990-1995) 45%, France (1990-1996) 28%, The Netherlands (1990-1995) 4%

3.1.2 General characteristics of industrial sources

The Emission Registration contains information on emissions, chimney heights and locations of individual (industrial) sources. This allows us to get an impression of the dispersion and load due to individual industrial sources. The following approach has been used:

- Sources of emissions are arranged according to the anticipated loads in the surroundings in terms of air concentrations with standard dispersion matrices in relation to a number of chimney height categories; the parameter[1] here is the *maximum* anticipated concentration.
- For 'high'-score sources, further information is obtained, wherever possible, by permit issuers, usually provincial authorities. In a number of cases the source and the emission circumstances could be better characterised on the basis of this supplementary information. This has usually led to a downward modification of the emission and in this way to a better insight into the necessity of possible monitoring.
- Finally, two source areas were selected for supplemental monitoring: one near the Hoogovens complex in Beverwijk and one in Rijnmond.

3.1.3 Arsenic

Using the OPS model, supplemental dispersion calculations were carried out for Aluminium Delfzijl, which resulted in a maximum anticipated contribution from this source of ca. 10 ng m^{-3} at a distance of less than 1 km from the source. The increase was on average 0.5 ng m^{-3} in an area of 5x5 km around the source. At 5 km from the source the contribution decreased by ca. 0.1 ng m^{-3}. In comparison, the Air Quality monitoring station, Kollumerwaard, nearby, measured an arsenic concentration of ca. 0.6 ng m^{-3} per year.

3.1.4 Cadmium

The outcomes of the standard dispersion matrices were not reason enough to make supplemental calculations with the OPS model.

3.1.5 Mercury

The outcomes of the standard dispersion matrices were not reason enough to make supplemental calculations with the OPS model.

3.1.6 Nickel

Using the OPS model, supplemental dispersion calculations were carried out for Aluminium Delfzijl, which resulted in a maximum anticipated contribution from this source of ca. 70 ng m^{-3} at a distance of less than 1 km from the source. The increase was on average 0.3 ng m^{-3} in an area of 5x5 km around the source. At 5 km from the source the contribution decreased by ca. 0.5 ng m^{-3}. In comparison, the nearby Air Quality monitoring station, Kollumerwaard[2], measured a nickel concentration of ca. 0.2 ng m^{-3} per year. Supplemental calculations were also made for the Rijnmond area, characterised by a number of nickel sources, especially in the refineries sector. Using the OPS model, and data on nickel emissions in the Rijnmond area in 1995, a dispersion calculation was carried out. See Figure 2 for the results. The substantial increase above the background levels remained confined to the province of Zuid-Holland**.**

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[1] See Appendix B for further information.

[2] Kollumerwaard is situated ca. 30 km west of Delfzijl.

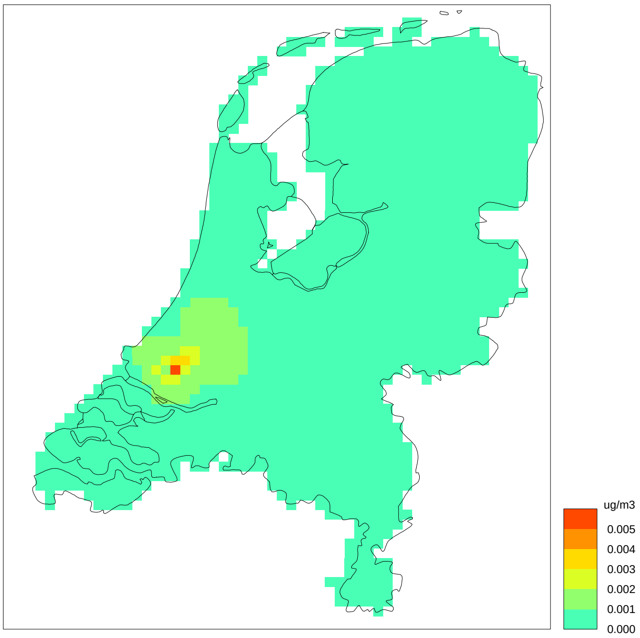

Figure 2 Contribution of nickel sources in the Rijnmond area to the nickel concentrations in the Netherlands in a grid of 5x5 km. The total concentration can be obtained by adding on a (background) concentration of 2 to 2.5 ng m^{-3}.

3.2 Measurements

For the sake of clarity a number of situation types will be distinguished: the background, industrial setting and remaining situations.

3.2.1 Background levels

The background level is described here as the level in situations outside the direct sphere of influence of the Netherlands' point or surface sources. The regional background level can be derived from the measurement results from the three regional LML locations and a number of locations in Zuid-Holland (see Table 3). Arsenic and cadmium background levels have been on the decrease for years, as shown in Figures 3 and 4. Due to a lack of long-range measurement series it is unknown if nickel levels have also been decreasing. An observable gradient showing decreasing concentrations from south to north has been found for arsenic and cadmium, reflecting the influence of the southern source areas of the Netherlands (Figures 5 and 6). For nickel the background levels across the Netherlands are more-or-less uniform.

Table 3 Yearly averages (ng m^{-3}) of arsenic, cadmium and nickel in air, 1998.

Station	Institution/ network(1)	Arsenic	Cadmium	Nickel
Biest Houtakker	RIVM-LML	1.1	0.39	2.3
Bilthoven	RIVM-LML	0.85	0.30	2.3
Kollumerwaard	RIVM-LML	0.64	0.21	2.1
Korendijkse Slikken	ZH	0.96	0.52	2.4
Hillegom	ZH	0.94	0.46	4.4
Bergambacht	ZH	0.92	0.44	3.3

1) RIVM-LML : RIVM/National Air Quality Monitoring Network
ZH : province of Zuid Holland

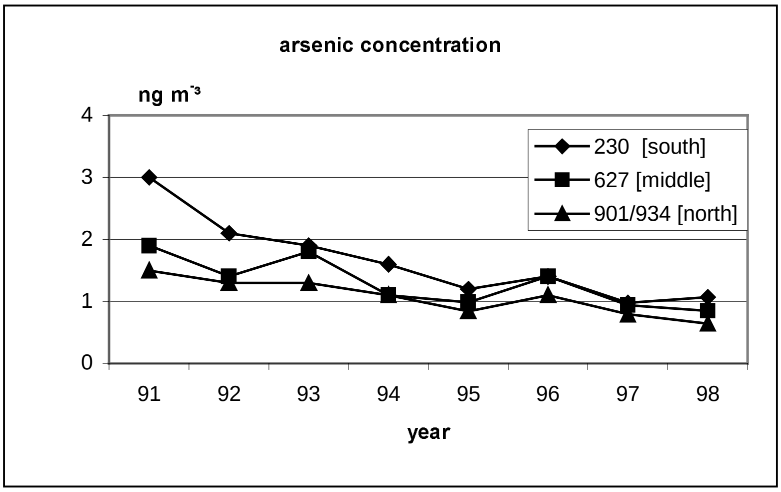

Figure 3 Development of arsenic concentration in air, 1991-1998.

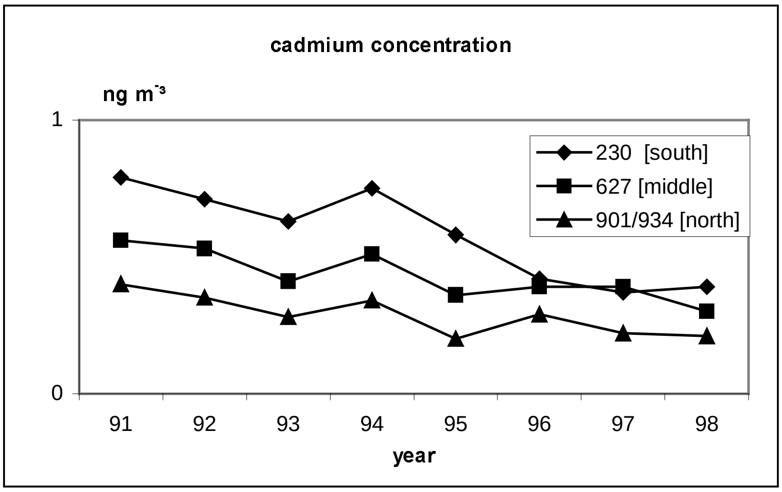

Figure 4 Development of cadmium concentration in air, 1991-1998.

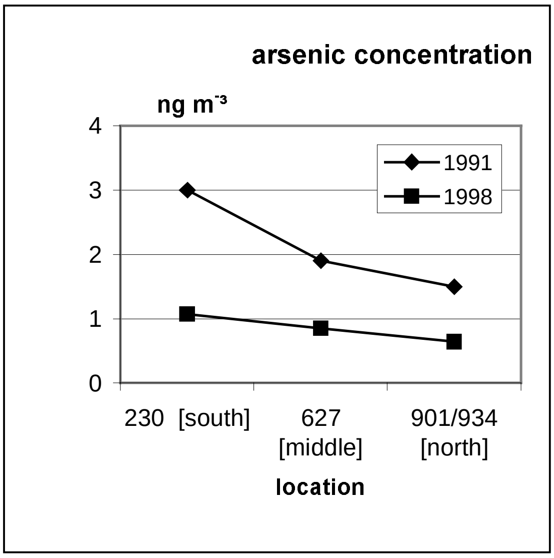

Figure 5 Gradient for the arsenic concentrations across the Netherlands 1991 and 1998.

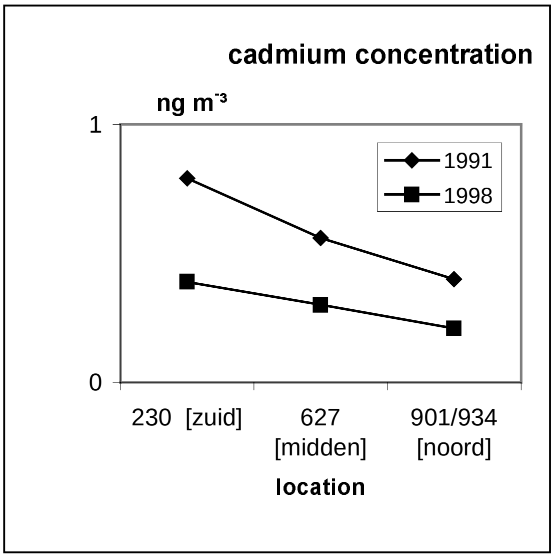

Figure 6 Gradient for the cadmium concentrations across the Netherlands, 1991 and 1998.

3.2.2 Industrial areas

In this section results will be presented of measurements with the explicit purpose of establishing the situation in the industrial areas and thus in the immediate vicinity of industrial sources. The two situations concerned here have to do with measurements for two locations: Zuid-Holland and the Rijnmond area, and Noord-Holland, and measurements taken by the province of Noord-Holland near the Hoogovens. In both cases the RIVM carried out monitoring in 1998 and 1999 in Schiedam and Beverwijk, respectively, to supplement the provincial monitoring. These results are summarised in Table 4. The concentrations of arsenic in air in the Netherlands are shown to be determined by the large-scale background level. Arsenic and cadmium levels rise slightly in the Rijnmond area, with increased levels of cadmium near the Hoogovens. Just as discussed in § 3.1.6 there is a clear rise in the nickel level anticipated in the Rijnmond area, as confirmed by the measurement results.

Table 4 Average concentration levels (ng m^{-3}) of arsenic, cadmium and nickel in industrial areas, 1998/99 (see Table 1 for the exact time periods).

Location	Institution/ network[1]	Arsenic	Cadmium	Nickel
Near the Hoogovens				
Beverwijk	RIVM	0.90	0.57	2.2
Wijk aan Zee[2]	NH	-	-	-
Rijnmond				
Schiedam	RIVM	1.1	0.38	16
Vlaardingen	RIVM-LML	1.2[3]	0.43[3]	4.7[3]
Hoek van Holland	ZH	0.98	0.57	8.6

[1] RIVM: supplemental measurements for the project 'Scanning and scouting measurements in air'
NH: province of Noord-Holland
RIVM-LML: RIVM/National Air Quality Network
ZH: province of Zuid-Holland

[2] All the measured concentrations are found below the analysis limit of the methods used (5 for arsenic and cadmium; 10 for nickel); reports of average concentrations are therefore of little value (see also Appendix D).

[3] Re-calculated concentration to be able to compare the other concentrations measured by RIVM; see also Appendix C.

RIVM has monitored arsenic and cadmium in Vlaardingen since the late 1980s. The development in yearly averages is found in Figure 7. Cadmium concentrations show a consistent decrease, and arsenic has shown a decreasing trend in the last few years.

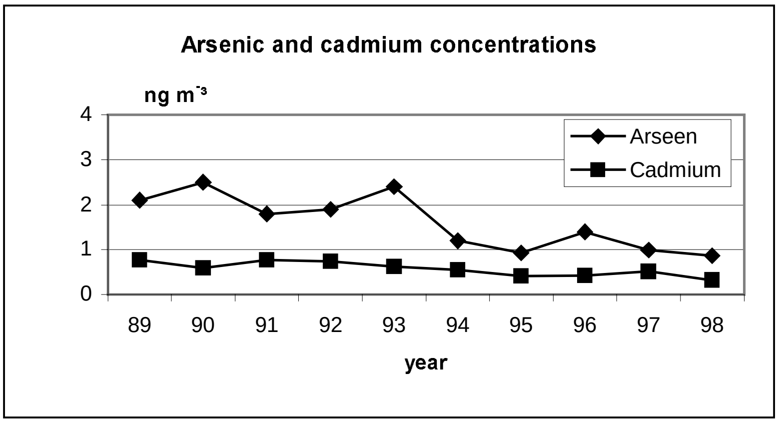

Figure 7 Yearly average for arsenic and cadmium concentrations in air in Vlaardingen, 1989-1998.

3.2.3 Other situations

It is unlikely that a local increase in arsenic, cadmium, mercury or nickel will occur outside the direct sphere of influence of individual sources or industrial areas. The measurements taken in Maastricht partly confirm this. The yearly average concentrations (1997) came to 0.7 ng m^{-3} for arsenic, 0.3 ng m^{-3} for cadmium and 1.1 ng m^{-3} for nickel.

3.2.4 Precipitation

Arsenic in precipitation is measured at a limited number of stations in the National Precipitation Chemistry Monitoring Network (LMRe); these are Rotterdam and the coastal station of Leiduin (replaced later by De Zilk). Up to 1995 the concentrations were almost always found under the analysis limit of the analysis method, i.e. 1.1 µg l^{-1}. Since 1996 a more sensitive method has been applied, resulting in concentrations of ca. 0.1 - 0.3 µg l^{-1}, corresponding to a wet deposition of ca. 80-240 µg arsenic m^{-2} j^{-1}.

Cadmium in precipitation has been measured for years at all the locations of the LMRe. Yearly average concentrations amount to 0.1-0.3 µg l^{-1}, which corresponds to a wet deposition of ca. 80-240 µg cadmium m^{-2} j^{-1}. In wet deposition there is generally no clear observable gradient.

Nickel is measured in precipitation at a limited number of stations in the LMRe; these are Rotterdam and the coastal station of Leiduin (replaced later by De Zilk). Up to 1995 the concentrations in De Zilk were almost always found below the analysis limit of the analysis method, i.e. 0.6 µg l^{-1}. Since 1996 a more sensitive method has been applied, resulting in concentrations of ca. 0.8 µg l^{-1}. In Rotterdam the yearly average concentration is ca. 1.2 µg l^{-1}, which means a wet deposition of ca. 1000 µg nickel m^{-2}.

In mid-1994 measuring *mercury* in precipitation was started at two stations in the LMRe: De Zilk and De Bilt. Since early 1998 monitoring has only been continued at De Zilk. Yearly average concentrations are in the order of 20-25 ng l^{-1} mercury per year at both locations. Where the average precipitation is 800 mm this will mean a wet deposition of ca. 20 µg mercury per m^2, corresponding well with the outcome of the model calculations (see §3.1.1).

4 Discussion

The volume of structural measurements for arsenic, cadmium and nickel in the Netherlands is not large. Mercury concentrations in air are not determined at all. Despite this, fairly reliable statements can be made about air quality in relation to these substances. *Arsenic* concentrations are in particular determined by their large-scale background levels, and measurements at regional stations do not give a very good picture of the levels in the Netherlands. The outcomes of the monitoring in the Rijnmond area and near the Hoogovens show hardly any rise when compared with the background levels. The increases in arsenic anticipated near the Hoogovens[1], as predicted by the model calculations, could not be confirmed by the measured levels.

Cadmium is measured both at background stations and in a number of industrial areas. This provides good insight into cadmium levels in the Netherlands and into the extent of locally enhanced levels. In the Rijnmond area, the cadmium level is slightly raised, while near the Hoogovens the increase amounts to 100% of the background level; here a local background concentration of 0.3 ng m^{-3} is assumed; the measured increase near the Hoogovens is, by the way, lower than could be anticipated from the model outcomes[2]

The air quality for *nickel* can be characterised by supplemental measurements from RIVM, allowing both the background level and the level of several Dutch source areas to be determined. The model calculations carried out for the sources in the Rijnmond area show a yearly average concentration of 7 to 8 ng m^{-3}. This in good agreement with the local monitoring, which shows concentrations in the order of 5-15 ng m^{-3}.

Statements on air quality for *mercury* are completely based on the outcomes of model calculations. The very large-scale dispersion pattern of mercury would probably have limited any extra information from monitoring.

The presence of arsenic, cadmium, mercury and nickel in the precipitation in the Netherlands is reasonably to well known. The arsenic and cadmium depositions show no spatial dependence across the Netherlands, although this cannot be said of mercury due to the limited number of monitoring stations. However, on the basis of the large-scale dispersion pattern and the lack of extensive sources of atmospheric mercury, it can be assumed that there would be little variation in the mercury deposition across the Netherlands. Only for nickel has a traditional increase of wet nickel deposition in Rijnmond been established. Just as the increase in nickel concentrations, this raised deposition is confined to the immediate vicinity of the source.

---

[1] The local contribution for the Beverwijk station is estimated at 0.6 ng As m^{-3} using model calculations.

[2] The local contribution for the Beverwijk station is estimated at 0.5 ng Cd m^{-3} using model calculations.

5 Conclusions

The presence of arsenic, cadmium, mercury and nickel in air and precipitation has been investigated in the project 'Scanning and scouting measurements for air'. Measurement data, limited supplemental monitoring efforts and the results of model calculations have made it possible to obtain good insight into air quality in the Netherlands with respect to these substances. The air quality related to these substances can be largely described on the basis of measurement results. Model calculations indicate that problem situations caused by industrial sources practically do not exist. For mercury, the description is largely based on model calculation outcomes. The calculated levels are, however, so low that mercury measurements would probably have not added any information.
The air quality for arsenic, cadmium, mercury and nickel in the Netherlands is summarised in Table 5. Comparing these levels with air quality objectives has taught us that the levels present in the Netherlands are usually found below these values[1]. Exceptions are cadmium and nickel, of which the Negligible Risk (NR) levels are exceeded.

Table 5 Average concentrations of arsenic, cadmium, mercury and nickel (ng m^{-3}) in air.

Situation	Arsenic	Cadmium	Mercury	Nickel
Background	0.6-1.1$^{1)}$	0.2-0.4$^{1)}$	2-3	2.2
Rijnmond area	1.0	0.4-0.5	-	5-15
Near Hoogovens	1.0^{2}	0.6	-	2.2$^{2)}$

$^{1)}$ Concentration gradient across the Netherlands; the lowest value is in agreement with the situation in the north of the Netherlands, the highest value with the south of the Netherlands.
$^{2)}$ Concentration is approximately comparable to the situational background level.

The presence of arsenic, cadmium, mercury and nickel in precipitation and the resulting deposition is summarised in Table 6.

Table 6 Average concentrations of arsenic, cadmium, mercury and nickel (ng m^{-3}) in precipitation in the Netherlands. Data given in µg l^{-1} (concentration) and µg m^{-2} j^{-1} (deposition)$^{1)}$.

Quantity	Arsenic	Cadmium	Mercury	Nickel
Concentration	0.10-0.30	0.10-0.30	0.025	0.80 [1.2]$^{2)}$
Deposition	60-240	80-240	20	640 [1000]^{2}

$^{1)}$ For the deposition calculation a yearly precipitation of 800 mm was used.
$^{2)}$ The (increased) values in the Rijnmond areas are in parentheses.

---

[1] *Arsenic:* WHO has not established a safe concentration for arsenic. The Netherlands employs an indicative MTR (Maximum Tolerated Risk) value of 500 ng m^{-3} and a target value of 5 ng m^{-3}.
Cadmium. WHO proposes a guide value of 1-5 ng m^{-3}; furthermore, the average concentration may not increase over time. For urban and industrial areas a value of 10-20 ng m^{-3} is allowed. The Netherlands employs an indicative MTR (Maximum Tolerated Risk) value of 25 ng m^{-3} and a NR (negligible risk) value of 0.25 ng m^{-3}.
Mercury. The Netherlands employs an indicative MTR (Maximum Tolerated Risk) value of 1 ng m^{-3} and a NR value of 10 ng m^{-3}; the limit value employed is 250 ng m^{-3} and the target value, 2.5 ng m^{-3}.
Nickel: WHO has not established a safe concentration for nickel. The Netherlands employs a limit value of 250 ng m^{-3} and a target value of 2.5 ng m^{-3}.

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We gratefully acknowledge the experience and expertise offered by the province of Noord-Holland and DCMR (advisory environmental service) Rijnmond in the inception phase of this report, Databases containing the measurements carried out by the provinces of Limburg, Noord-Holland and by DCMR Rijnmond were of value in the reporting phase of the investigation. Measurement data were kindly made available by the above-mentioned institutions. Information on a number of industrial sources was provided by the provinces of Gelderland, Groningen, Limburg and Noord-Holland, as well as the Hoogovens. Ruth de Wijs-Christensen is thanked for the editorial assistance.

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Appendix A Mailing list

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27	Bibliotheek RIVM
28-43	Bureau Rapportenbeheer
44-70	Reserve exemplaren

Appendix B Standard dispersion calculations

Standard dispersion matrices have been used to estimate the maximum concentration due to individual industrial sources. These standard dispersion matrices were calculated with the Operational Priority Substances Model OPS, version 2.1.1, for a limited number of chimney heights (10, 25, 50, 100 m), an emission of 1 g s^{-1} (≈32 tons a^{-1}) and a heat content of 0 MW. This serves as a quick method to estimate concentration levels from a specific source. Concentrations could then be related to limit values, for example, to see if further research would be necessary. The advantage of this approach is that only a limited number of dispersion calculations have to be carried out. Figure B1 gives an example of a dispersion matrix for a source with a chimney height of 50 m. As the model is linear for the ratio emission/concentration, it is possible to make an easy calculation of the concentration dispersion (and maximum concentration) for each emission strength.

224	232	240	241	251	244	241	246	270	276	291	316	346	387	390	424	444	458	488	490	485	470
241	246	249	262	260	268	244	247	268	278	288	320	346	416	440	462	492	502	510	525	513	489
259	263	272	269	263	272	263	262	281	314	306	337	384	427	478	514	541	545	548	533	503	482
277	286	283	286	288	281	300	292	300	333	337	367	425	460	524	562	593	602	576	553	533	475
305	300	302	306	295	300	313	322	319	353	351	387	451	502	567	611	640	633	589	570	523	512
329	320	326	314	317	335	343	358	340	355	362	403	490	542	615	663	667	619	603	557	548	527
341	349	343	340	360	362	363	366	379	364	389	413	469	581	658	688	639	594	596	582	559	531
335	368	360	369	390	392	400	391	386	377	411	486	622	763	677	635	601	611	602	580	553	522
344	348	354	350	404	419	425	425	377	344	330	444	622	785	691	575	596	597	581	561	534	508
352	359	360	358	352	342	320	425	357	217	135	778	815	600	665	533	559	596	588	568	539	515
348	358	362	362	359	346	319	256	139	420	848	694	1023	515	624	555	583	586	575	559	538	509
339	334	340	342	339	327	297	232	142	240	791	● 1439	1070	390	530	516	543	549	545	528	506	489
317	326	331	349	429	437	492	487	417	300	458	759	380	556	658	479	505	507	501	488	477	454
347	361	355	369	397	405	390	416	390	308	202	246	313	527	574	565	579	571	471	462	445	428
334	345	348	347	352	355	333	323	308	356	267	320	341	390	493	509	516	514	498	469	416	405
317	328	336	317	310	305	305	265	259	227	259	306	309	345	367	464	466	465	454	421	407	432
293	301	304	288	270	280	250	228	271	236	264	303	298	323	343	359	426	418	388	386	371	394
268	268	272	267	266	253	221	224	250	240	259	290	287	303	319	328	341	362	361	349	370	364
244	243	251	241	237	224	220	239	214	234	251	273	272	282	296	303	309	316	328	351	342	327
223	227	220	217	208	201	219	234	211	227	239	260	279	264	273	281	286	288	330	323	313	304
206	204	199	195	188	192	205	217	210	220	228	246	260	243	256	262	268	254	252	295	287	282
189	189	181	177	177	184	195	183	194	201	220	232	234	229	239	229	232	233	233	230	269	251

grid cell dimensions : 50 m
number of grid points : 22x22
co-ordinates upper left corner : -525 m, 525 m
average concentration : 384 ng m^{-3}

Figure B1 Example of a standard dispersion matrix for a source with an emission strength of 1g s^{-1} and a chimney height of 50 m. Numbers refer to yearly averaged concentrations in ng m^{-3}. Source location is given by ● . Maximum concentration is underlined.

Tables B1 up to and including B4 give the sources that were first evaluated by standard dispersion matrices and for which it was concluded that high levels could possibly occur. This table also shows first-order estimates for the maximum concentration and the distance D of the maximum. Based on these results, it was concluded that for the Hoogovens (Steelworks) and for the heavily industrialised Rijnmond area, supplementary dispersion calculations on a more spatially detailed level has to be carried out.

Table B1 First-order estimates of maximum arsenic concentrations.
Input data from Emission Registration 1995.

Source	Location	C_{max} (ng m^{-3})	D (in m)[1)]	Remark
Aluminium Delfzijl	Delfzijl	2000	<100	See discussion in § 3.1.3
Outocompu	Gelderland	40	<100	Emission substantially lower[2)]
Hoogovens	Beverwijk	30	<100	Supplementary measurements
DSM	Geleen	20	<100	Emission substantially lower [3)]

[1)] The validity of the model does not extend over distances of less than 100 m from the source.
Therefore if the model calculates the maximum concentration in the first grid element, '<100' is given as the location of the maximum.
[2)] Information given by the issuer of the permit, the province of Gelderland.
[3)] Information given by the issuer of the permit, the province of Limburg.

Table B2 First-order estimates of maximum cadmium concentrations.
Input data from Emission Registration 1995.

Source	Location	C_{max} (ng m^{-3})	D (in m)[1)]	Remark
Outocompu	Gelderland	200	<100	Emission substantially lower[2)]
Budelco	Budel	100	<100	
Aluminium Delfzijl	Delfzijl	20	<100	
Hoogovens	Beverwijk	10	<100	Supplementary measurements

[1)] The validity of the model does not extend over distances of less than 100 m from the source.
Therefore if the model calculates the maximum concentration in the first grid element, '<100' is given as the location of the maximum.
[2)] Information given by the issuer of the permit, the province of Gelderland

Table B3 First-order estimates of maximum mercury concentrations.
Input data from Emission Registration 1995.

Source	Location	C_{max} (ng m^{-3})	D (in m)[1)]	Remark
Solvay	Limburg	300	<100	
DSM	Geleen	200	<100	

[1)] The validity of the model does not extend over distances of less than 100 m from the source.
Therefore, if the model calculates the maximum concentration in the first grid element, '<100' is given as the location of the maximum.

Table B4 First-order estimates of maximum nickel concentrations.
Input data from Emission Registration 1995.

Source	Location	C_{max} (ng m^{-3})	D (in m)[1]	Remark
Aluminium Delfzijl	Delfzijl	1000	<100	See discussion in § 3.1.6
Nerefco	Pernis	1000	750	Supplementary measurements
Bakker	Z Holland	700	<100	
Henckels	Overijssel	700	<100	
DSM	Geleen	400	<100	
UNI Mills	Z Holland	400	<100	

[1] The validity of the model does not extend over distances of less than 100 m from the source.
Therefore, if the model calculates the maximum concentration in the first grid element, '<100' is given as the location of the maximum.

Models
The standard dispersion matrices, for which the results have been presented in Chapter 3, were calculated with the Operational Priority Substances Model, version 2.1.1. Data on emissions, chimney heights and heat content for individual industrial sources are taken from the Emission Registration 1995. Calculations were carried out with long-term averaged meteorological data (1960-1990).

Appendix C Sampling and analysis of metals

Measurements/RIVM

The sampling of arsenic, cadmium and nickel at the locations of Beverwijk (555) and Schiedam (427) was carried out with the Partisol-Plus 2025 with PM10 inlet, manufactured by Rupprecht & Patashnick. A Teflon filter from Gelman Sciences R2PJ047 was used. Sampling was continuous, with a sampling time of 24 hours. Sampled volume was 24 m^3.

Biest-Houtakker, Bilthoven, Kollumerwaard and Vlaardingen are locations in the Dutch Air Quality Monitoring Network where metals are re-sampled on a routine basis with medium volume samplers. A Whatman 42 filter is used. Sampling time is 24 hours; total volume is approx. 8 m^3. Filters were analysed on a routine basis for arsenic and cadmium, and on this occasion also for nickel. Results obtained with two different sampling methods were used in this research. To facilitate the comparability of the two sampling methods, the standard monitoring network equipment was used at the location of Beverwijk. The Laboratory for Inorganic Analysis at RIVM carried out the analysis. Filters were destructed in a microwave oven with 1.5 M nitric acid for 1½ hours. Analysis was done by ICP-MS. Field and laboratory blanks were taken on a regular basis during the investigation.

The results of the measurements carried out in parallel in Beverwijk are shown in Figure C1 to C3. The regression lines shown in these figures can be used to re-calculate the Whatman filter results and thereby make them comparable to the Teflon filter results. This procedure has been used to obtain the results shown in Table 4.

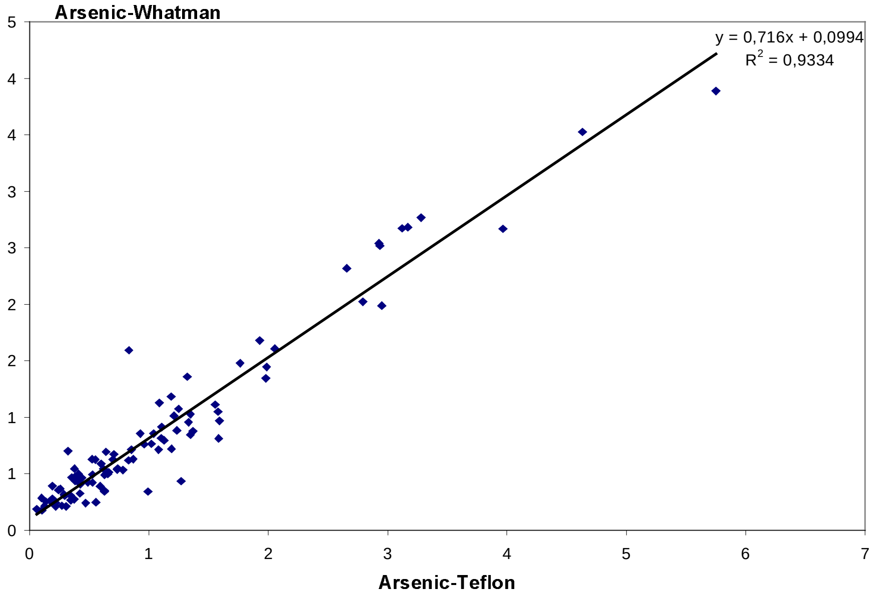

Figure C1 Comparison of arsenic concentrations using Teflon and Whatman filters.

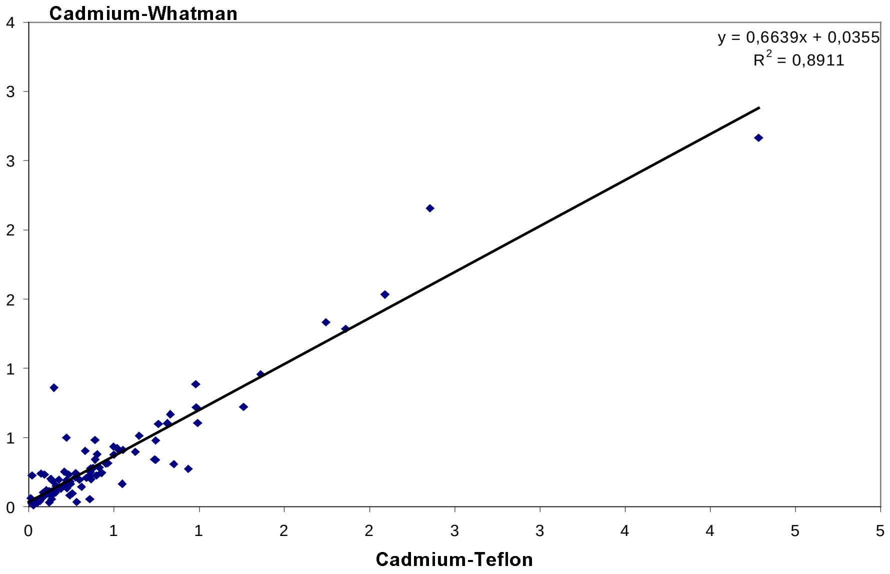

Figure C2 Comparison of cadmium concentrations using Teflon and Whatman filters.

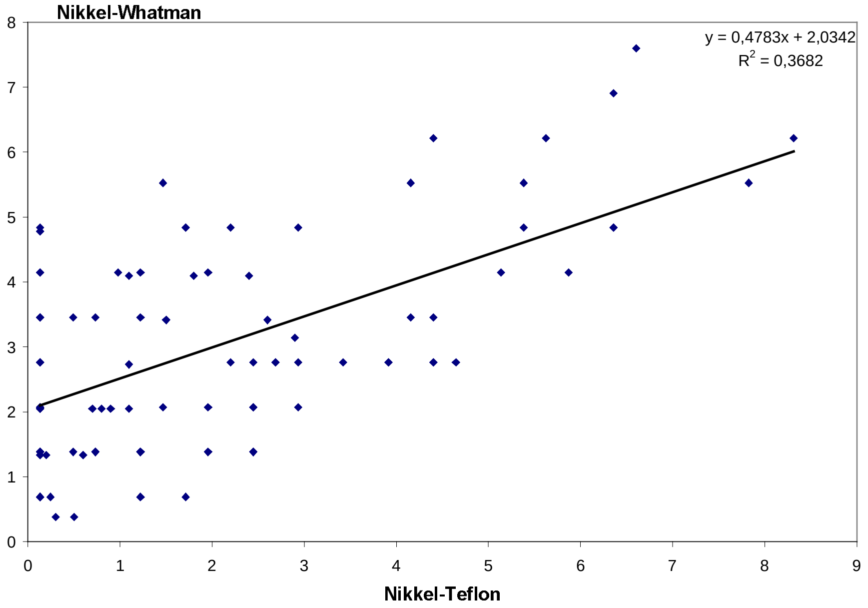

Figure C3 Comparison of nickel concentrations using Teflon and. Whatman filters.

Measurements/Limburg

Sampling was carried out with a High Volume Sampler, type GUV-15 H, manufactured by Anderson Samplers Inc./ General Metal Works Inc. The device had an PM-10 size selective sampling inlet, model G 1200. Up to 1994, Whatman 41 type filters made of glass fibre had been used.. Since 1995 QMA filters made of quartz fibre have been used. Sampling took place every 5 days; the sampling period lasted 24 hours.. Filters were analysed in the provinces (Limburg) with ICP.

Measurements/Noord-Holland
Sampling was carried out with Sierra/Andersen PM10-model 1200-high volume samplers with a PM10 inlet. Whatman QMA quartz fibre filters were used. Sampling period was 24 hours.

Measurements/Zuid-Holland
Sampling was carried out with the Digital DH-80 high volume sampler. Filters from Schleier&Schull, type GF-50, and Whatman QM-A type quartz fibre, were used. The flow was approximately 32 $m^3 h^{-1}$. Measurement results were corrected for blanks. Average blank values were calculated using the results of 23 blank filters. Filters were destructed in a microwave oven in a mixture of concentrated nitric acid and hydrochloric acid. Analysis was done with atomic emission spectrometry met ICP-USV. Analysis was continued with ICP-AES according to NEN 6426. A hydride technique was used for arsenic.

General remarks on sampling
A summary of the most important characteristics of the different sampling methods is given in Table C1.

Table C1 Characteristics of the sampling methods by the different institutions/network.

Institution/network	Flow ($m^3 h^{-1}$)	Period	Sampling	Remark
Province of Limburg	-[1]	1 day	Quartz fibre	PM10 inlet
Province of Noord-Holland	68	1 day	Quartz fibre	PM10 inlet
Province of Zuid-Holland[2]	32	1 day	Quartz fibre	
RIVM-LML	0.1	1 day	Whatman 42	
RIVM[3]	1	1 day	Teflon	PM10 inlet

[1] Information not available.

[2] Sampling by DCMR (environmental advice) Rijnmond.

[3] Supplementary sampling within the project ‘Scanning and scouting measurements for air’.

Appendix D Measurement results

N.B.: The measurement results in this Appendix will be presented as they were reported by the institutions/networks. Only blank corrections were made.

Table D1 Arsenic concentrations in air (ng m^{-3}) at locations of the Dutch National Air Quality Monitoring Network, 1991-1998.

Location	1991	1992	1993	1994	1995	1996	1997	1998
Biest-Houtakker	3.0	2.1	1.9	1.6	1.2	1.4	0.98	1.1
Bilthoven	1.9	1.4	1.8	1.1	0.99	1.4	0.94	0.85
Kollumerwaard	1.5	1.3	1.3	1.1	0.84	1.1	0.79	0.64
Vlaardingen	2.5	1.8	2.4	1.2	0.93	1.4	0.99	0.86
Beverwijk	.	.	.	.	.	.	.	0.90$^{1)}$
Schiedam	.	.	.	.	.	.	.	1.1$^{1)}$

$^{1)}$ Averaged over the period of September 1998 to June 1999.

Table D2 Arsenic concentrations in air (ng m^{-3}) at regional network locations, 1992-1998.

Location	1991	1992	1993	1994	1995	1996	1997	1998
Province of Noord-Holland								
De Rijp	.	.	.	.	.	.	.	<5
Wijk aan Zee	.	.	.	.	.	.	.	<5
Province of Zuid-Holland								
Korendijkse Slikken	.	.	.	.	.	2.0	1.3	0.96
Hillegom	.	.	.	.	.	1.7	1.4	0.94
Bergambacht	.	.	.	.	.	1.2	1.4	0.92
Westland	.	.	.	.	.	1.9	1.3	.
Hoek van Holland	.	.	.	.	.	.	.	0.98
Province of Limburg								
Maastricht	.	1.0	1.9	1.5	1.5	1.8	0.7	.

Table D3 Cadmium concentrations in air (ng m^{-3}) at locations of the Dutch National Air Quality Monitoring Network, 1991-1998.

Location	1991	1992	1993	1994	1995	1996	1997	1998
Biest-Houtakker	0.79	0.71	0.63	0.75	0.58	0.42	0.37	0.39
Bilthoven	0.56	0.53	0.41	0.51	0.36	0.39	0.39	0.30
Kollumerwaard	0.40	0.35	0.28	0.34	0.20	0.29	0.22	0.21
Vlaardingen	0.59	0.74	0.62	0.55	0.41	0.42	0.51	0.32
Beverwijk	.	.	.	.	.	.	.	0.57$^{1)}$
Schiedam	.	.	.	.	.	.	.	0.38$^{1)}$

$^{1)}$ Averaged over the period of September 1998 to June 1999.

Table D4 Cadmium concentrations in air (ng m^{-3}) at locations of regional networks, 1992-1998.

Location	1991	1992	1993	1994	1995	1996	1997	1998
Province of Noord-Holland								
De Rijp	.	.	.	.	.	.	.	<5
Wijk aan Zee	.	.	.	.	.	.	.	<5
Province of Zuid-Holland								
Korendijkse Slikken	.	.	.	.	.	0.54	0.46	0.52
Hillegom	.	.	.	.	.	0.41	0.33	0.46
Bergambacht	.	.	.	.	.	0.34	0.40	0.44
Westland	.	.	.	.	.	0.54	0.45	.
Hoek van Holland	.	.	.	.	.	.	.	0.57
Province of Limburg								
Maastricht	1.0	1.0	1.3	2.3	1.1	0.7	0.3	.

Table D5 Nickel concentrations in air (ng m^{-3}) at locations of the Dutch National Air Quality Monitoring Network, 1991-1998.

Location	1991	1992	1993	1994	1995	1996	1997	1998
Biest-Houtakker	.	.	.	.	.	.	.	2.3
Bilthoven	.	.	.	.	.	.	.	2.3
Kollumerwaard	.	.	.	.	.	.	.	2.1
Vlaardingen	.	.	.	.	.	.	.	4.3
Beverwijk	.	.	.	.	.	.	.	2.2(1)
Schiedam	.	.	.	.	.	.	.	16(1)

(1) Averaged over the period September 1998 to June 1999.

Table D6 Nickel concentrations in air (ng m^{-3}) at regional network locations, 1992-1998.

Location	1991	1992	1993	1994	1995	1996	1997	1998
Province of Noord-Holland								
De Rijp	.	.	.	.	.	.	.	<10
Wijk aan Zee	.	.	.	.	.	.	.	<10
Province of Zuid-Holland								
Korendijkse Slikken	.	.	.	.	.	5.8	3.0	2.4
Hillegom	.	.	.	.	.	4.7	2.5	4.4
Bergambacht	.	.	.	.	.	1.5	2.6	3.3
Hoek van Holland	.	.	.	.	.	.	.	8.6
Westland	.	.	.	.	.	9.6	8.4	.
Province of Limburg								
Maastricht	.	.	.	.	.	.	1.1	.

Table D7 Arsenic concentrations in precipitation (µg l^{-1}) at locations of the Dutch National Air Quality Monitoring Network, 1991-1998.

Location	1993	1994	1995	1996	1997	1998
De Zilk	.	<1.1	<1.1	0.30	0.07	.
Rotterdam	<1.1	<1.1	<1.1	0.22	0.22	.

Table D8 Cadmium concentrations in precipitation (µg l^{-1}) at locations of the Dutch National Air Quality Monitoring Network, 1991-1998.

Location	1993	1994	1995	1996	1997	1998
Beek	0.15	0.25	0.30	0.26	0.15	.
De Bilt	0.13	0.18	0.30	0.26	0.12	0.12
De Zilk	.	.	0.98	0.13	0.11	0.12
Gilze-Rijen	0.13	0.15	0.12	0.13	0.15	0.08
Kollumerwaard	0.16	0.12	0.71	0.22	0.24	0.15
Rotterdam	0.18	0.20	0.13	0.19	0.11	0.15

Table D9 Nickel concentrations in precipitation (µg l^{-1}) at locations of the Dutch National Air Quality Monitoring Network, 1991-1998.

Location	1993	1994	1995	1996	1997	1998
De Zilk	.	0.88	0.88	0.59	0.82	.
Rotterdam	1.5	1.1	1.2	1.2	1.2	.

Table D10 Mercury concentrations in precipitation (ng l^{-1}) at locations of the Dutch National Air Quality Monitoring Network, 1991-1998.

Location	1993	1994	1995	1996	1997	1998
De Bilt	.	.	24	28	23	.
De Zilk	.	.	25	27	17	16