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RIVM report 729999 001

Assessment of air quality for Polycyclic Aromatic Hydrocarbons in the Netherlands

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Abstract

The presence of polycyclic aromatic hydrocarbons (PAHs), and then especially the PAH indicator benzo[a]pyrene (B[a]P), 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. The background level of B[a]P is estimated at 0.05 – 0.15 ng m^{-3} on a yearly average. Increased PAHs and B[a]P concentrations are expected in the vicinity of a number of industrial sources and in urban surroundings. B[a]P concentrations near the Hoogovens (Steelworks) measure 0.2 to 0.5 ng m^{-3}, depending on their position in relation to the source complex. The urban background level comes to 0.2 to 0.4 ng m^{-3}; on roads yearly averages of up to 0.7 ng m^{-3} can occur. Model calculations show results in streets of ca. 20 km in length in which the limit value of 1 ng m^{-3} is exceeded. This is not confirmed by measurement. Further research will be needed to show the cause of the apparent discrepancy.

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 PAK in Nederland. In dit rapport worden de resultaten van het onderzoek dat is uitgevoerd naar de luchtkwaliteit van deze stof, besproken. Hierbij is de aandacht vooral gericht op het voor PAK als gidsstof beschouwde benzo[a]pyreen. 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 het achtergrondniveau, over de situatie nabij een belangrijke industriële PAK bron en vooral over de situatie in steden.
De luchtkwaliteit voor PAK kan getoetst worden aan de huidige Nederlands grens- en richtwaarde voor benzo[a]pyreen van 0,5 resp. 1,0 ng benzo[a]pyreen per m^3 als jaargemiddelde.

De meetresultaten kunnen gebruikt worden om gemiddelde niveaus in een aantal situaties aan te geven. Zo wordt het achtergrondniveau van benzo[a]pyreen geschat op 0,05-0,15 ng m^{-3} als jaargemiddelde. Verhoogde PAK- en benzo[a]pyreenconcentraties worden verwacht in de nabijheid van een aantal industriële bronnen en in de stedelijke omgeving. In de nabijheid van de Hoogovens bedragen de benzo[a]pyreenconcentraties, afhankelijk van de positie ten opzicht van het bronnencomplex, 0,2 tot 0,5 ng m^{-3} als jaargemiddelde. Het stedelijke achtergrondniveau bedraagt ca. 0,2 tot 0,4 ng m^{-3}; in straten kunnen jaargemiddelde concentraties voorkomen tot 0,7 ng m^{-3}. Berekeningen met modellen geven als uitkomst dat in straten langs ca. 20 km weglengte overschrijding van de grenswaarde van benzo[a]pyreen van 1 ng m^{-3} zou kunnen optreden. Dit wordt niet door de metingen bevestigd. Nader onderzoek zal moeten uitwijzen waardoor deze ogenschijnlijke discrepantie wordt veroorzaakt.

Summary

Support for 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 PAHs in the Netherlands, focusing especially on the PAH indicator, benzo[a]pyrene. An important aspect will be to make an inventory of the potential problem situations.

This investigation made primary use of available information to characterise air quality. The investigation is supplemented with limited monitoring efforts for the situations in which research has shown the present knowledge to be insufficient. This monitoring effort was focused on improving the knowledge on background levels and on situations near an important industrial PAH source, especially the situation in cities. Air quality for PAHs can be tested against the current Dutch limit and average yearly guide values of 0.5 and 1.0 ng m^{-3} for benzo[a]pyrene and benzo[a]pyrene, respectively.

Measurement results can be used to indicate average levels in a number of situations. For instance, the background level of benzo[a]pyrene is estimated at 0.05 – 0.15 ng m^{-3} on a yearly average. Increased PAH and benzo[a]pyrene concentrations are expected in the vicinity of a number of industrial sources in urban surroundings. Benzo[a]pyrene concentrations near the Hoogovens (Steelworks) measure 0.2 to 0.5 ng m^{-3}, depending on their position in relation to the source complex. The urban background level comes to 0.2 to 0.4 ng m^{-3}; on roads, yearly averages of up to 0.7 ng m^{-3} can occur. Model calculations show results in streets of ca. 20 km in length in which the limit value of 1 ng m^{-3} is exceeded. This is not confirmed by measurement. Further research will be needed to show the cause of the apparent discrepancy.

Contents

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 particulates 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 (focused especially on benzo[a]pyrene). An important aspect is to inventory situations where raised levels of substances might occur. This report (729999 001) will deal with PAHs, while in a separate report (729999 002), arsenic, cadmium, mercury and nickel are discussed.

Chapter 2 discusses the general set-up and the investigation, as well as research methods and materials, 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 PAHs (sources, emissions, effects) will not be discussed in this report; please refer to the Basic Document for PAH (Slooff *et al.*, 1989). Appendix B offers a list of a number of physico-chemical properties of the most important PAHs.

2 Material and methods

2.1 Research outline

The starting point for making this assessment was that use should be made of information already available, especially that derived from measurements. Eventually, this information could be supplemented with the results of model calculations. Dispersion models were to be used to detect potential problem situations in the vicinity of industrial sources. The assessment would be concluded by examining the remaining gaps in the information 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 monitoring activities presented in §2.3. These were used by the RIVM to define a monitoring programme, which will be outlined at the end of §2.3.

Many PAHs show harmful effects, which, however, differ greatly in severity. This report will focus on heavier PAHs[1], especially on benzo[a]pyrene; the reason for this is partly sampling problems with the lighter PAHs. The focus on benzo[a]pyrene is also inspired by the fact that benzo[a]pyrene is supposed to be an indicator of PAH.

2.2 Model calculations

The model calculations, with which concentration levels due to industrial sources have been estimated, were carried out using the Operational Priority Substances model. Van Jaarsveld (1990, 1995) has described this model. Input data for the model on emission strength, chimney height and heat content were taken from the Dutch Emission Registration. Further details can be found in Appendix C. Data on exceedances of benzo[a]pyrene limit values in cities were derived from calculations using the CAR model[2].

2.3 Measurements

The inventory of operational and recently performed monitoring activities:

- Measurements by the provincial authority of Noord-Holland were taken both near the Hoogovens Steelworks, and Schiphol airport, as well as at a background station in the middle of the province of Noord-Holland. These are 24-hour samples, taken every eight days.
- Measurements by the DCMR Milieudienst (advisory environmental service*)* Rijnmond were taken at urban locations in Rotterdam. These are 24-hour samples, taken every six days.
- Measurements were taken by the provincial authority of Zeeland near an industrial source in Sluiskil. These are 24-hour samples, taken every three days.
- Measurements by OMEGAM at background station De Zilk were taken for a number of months in 1998. These were continuous measurements taken within the framework of an international comparison research co-ordinated by OSPAR (Holz, 1999).

A summary of PAHs reported by the aforementioned organisations/networks is given in Appendix D. The measurement results were obtained using different sampling procedures, which hampers a direct comparison of measurement results. The resulting problems will be discussed in Appendix E. Measurement, sampling and analysis methods are discussed in more detail in this Appendix.

[1] This refers to PAH which are sampled on a filter with an efficiency of 90% or more: benzo[a]pyrene, benzo[b]fluoranthene, benzo[ghi]perylene, benzo[k]fluoranthene, dibenzo[ah]anthracene, indeno[123-cd]pyrene (Baek *et al*., 1991, Harrison *et al*., 1996; Smith & Harrison, 1996; see also Appendix E).
[2] CAR stands for Calculation of Air pollution by Road traffic; see Eerens *et al.* (1993) for details.

Some additional information on the occurrence of PAHs in the urban environment, as well as at background stations, can be derived from earlier research by RIVM (Van Velze, 1996). The inventory of monitoring activities showed only a limited number of measurements in the urban environment, especially with respect to situations with (heavy) traffic, for which enhanced PAH levels are to be expected (Slooff *et al.*, 1989). Furthermore, the model calculations showed (see further in § 3.1) that one industrial source could give rise to a potentially problematic situation. Finally, the background level in the southern part of the Netherlands is not well known. The abovementioned observations have led to the definition of additional PAH measurements by RIVM at five locations: three urban sites, one industrial and one background site.

The interest in PAHS is mainly inspired by their harmful influence on human health. Since long-term exposure is of interest, the RIVM monitoring strategy focuses on the determination of (long-term) average PAH levels. A relatively long sampling period of one week could therefore be chosen. A characterisation of measurements and monitoring sites of all the organisations is given in Table 1. Figure 1 shows the locations of the monitoring sites.

In the past some evidence has been found that in certain situations (individual) PAHs have shown a strong correlation with black smoke (Van Velze, 1996; Coleman, 1999). Therefore black smoke measurements have been carried out in parallel with the PAH measurement in the RIVM monitoring programme. The same approach was followed by DCMR at their monitoring sites. Black smoke measurements were performed in the same way as in the Dutch National Air Quality Monitoring Network (Van Elzakker & Buijsman, 1999). However, discussion of the results of this black smoke subproject is beyond the scope of this publication.

Figure 1 Locations with operational or recent PAH measurements
[□: background ▲: industrial ❍: urban ■: other].

Table 1 PAH measurements reported here.

Monitoring site	Type[1)]	Time period
DCMR Environmental Institute Rijnmond		
Rotterdam-Vasteland	Urban background	1/1991-12/1998
Rotterdam-Statenweg	Traffic[2)]	1/1994-12/1995
OMEGAM		
De Zilk	Regional background	9/1998-10/1998
Province of Noord-Holland		
Wijk aan Zee-Verlengde Voorstraat	Industrial source	1/1991-12/1995
Wijk aan Zee-Banjaert	Industrial source	1/1996-12/1998
Ijmuiden	Industrial source	1/1991-12/1998
De Rijp	Regional background	1/1991-12/1998
Badhoevedorp	Surface source/traffic	1/1995-12/1998
Hoofddorp	Surface source	1/1995-12/1998
Oude Meer	Surface source	1/1994-12/1998
Province of Zeeland		
Sluiskil	Industrial source	1/1995-12/1996
National Institute of Public Health and the Environment (RIVM)		
Apeldoorn-Stationsstraat (728) [3)]	Traffic[4)]	2/1992- 4/1992[7)]
Beverwijk (555)	Industrial source	10/1998- 6/1999
The Hague -Rebequestraat (404)	Urban background	2/1992- 4/1992[7)]
Utrecht-Wittevrouwenstraat (637)	Traffic [5)]	10/1998- 6/1999
Utrecht-Erzeijstraat (639)	Traffic [6)]	10/1998- 6/1999
Utrecht-UB tuin (640)	Urban background	9/1998- 6/1999
Vredepeel (131)	Regional background	9/1998- 6/1999
Westmaas (437)	Regional background	2/1992- 4/1992[7)]

[1)] Type according to organisation/network.
[2)] Monitoring site located next to a roadway with six lanes; approx. 40,000 vehicles a day, distance to the nearest roadside is several metres and to the separation strip approximately 15 metres.
[3)] Station number in the Dutch National Air Quality Monitoring Network is given in parentheses.
[4)] Approximately 16,000 vehicles a day; distance to the nearest roadside is 4 metres; distance to the road axis is 10 metres; contribution of heavy traffic is 10%.
[5)] Approx. 15,000 vehicles a day; distance to the nearest roadside is 2 metres; distance to the road axis is 5 metres; contribution of heavy traffic is 10%.
[6)] Approx. 15,000 vehicles a day; distance to the nearest roadside is 1 metre; distance to the road axis is 6 metres; contribution of heavy traffic is 10%.
[7)] Limited number of samples.

3 Results

3.1 Model calculations

3.1.1 Introduction

The heavier PAHs are mostly particle-bound, attaching mainly to particles with an aerodynamic diameter of less than 4 µm (Vaek & Cauwenberghe, 1985), which gives PAHs a large-scale dispersion pattern. In the model calculations for the European scale, last carried out for 1989 (RIVM, 1994; Baart *et al*., 1995), the background level in the Netherlands was shown to be about 0.4 ng m^{-3}. Furthermore, the calculations showed the concentration in air of most of the PAHs in the Netherlands determined by Dutch sources to be 30-50% (benzo[a]pyrene ca. 40%) and by foreign sources to be 70-50%. However, these statistics are plagued with large uncertainties. This is due, on the one hand, to uncertainty in the emissions and, on the other, to hardly being able to test the results of model calculations due to lack of (structural) monitoring.

3.1.2 General characterisation of industrial sources

The Emission Registration contains information on emissions[1], chimney heights and locations of individual (industrial) sources. This gives one 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 expected loads in the surroundings in terms of air concentrations with standard dispersion matrices in relation to a number of chimney height categories; the parameter[2] here is the *maximum* expected 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, a calculation could be made for one source, the Hoogovens (Steelworks) in IJmuiden, using the OPS dispersion model, whereby information on air concentrations in the surroundings of this source became available.

3.1.3 The IJmuiden Hoogovens (Steelworks) in Noord-Holland

The Steelworks appears to be the only source for which the outcomes of the model calculations indicate the continuous presence of benzo[a]pyrene concentrations close to guide or limit values (0.5 and 1 ng m^{-3}, respectively). The province of Noord-Holland has for years carried out PAH measurements northwest and southeast of this source. This was followed by a decision in consultation with the province to install a supplementary RIVM monitoring station northeast of the Steelworks complex. This collective strategy allows diverse dispersion circumstances to be measured, including the most often occurring meteorological situation, i.e. the southwest wind. The results of these measurements will be discussed in section 3.2. Calculated PAH and benzo[a]pyrene concentrations are presented in Figure 2.

[1] PAH emissions are included in the Emission Registration as the 10 VROM PAH, see also Appendix D.
[2] See Appendix C for further information.

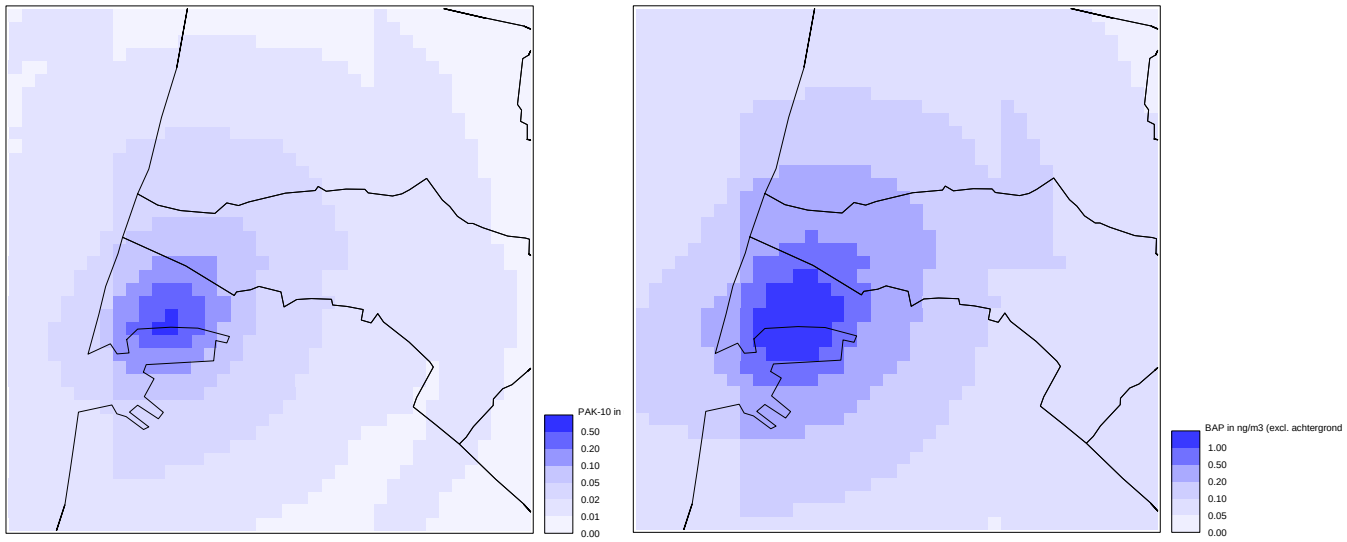

Figure 2 PAH concentrations (as the 10 VROM PAH, in µg m^{-3}) on the left and concentrations of benzo[a]pyrene (in ng m^{-3}) on the right as found near the Steelworks. Concentrations were calculated with the OPS model on the basis of emission data from the Emission Registration for 1996. The benzo[a]pyrene concentrations were obtained by multiplying the PAH concentrations by a factor of 0.007 (see further information in section 4). The cited concentrations exclude the background contribution, estimated for benzo[a]pyrene at 0.00005-0.00015 µg m^{-3} (0.05-0.15 ng m^{-3}). Scale: 1 cm≈2 km.

3.1.4 Streets

A general impression of benzo[a]pyrene concentrations in streets can be obtained by model calculations, for example, using CAR. This is a simple parameterised model requiring data on emission factors, traffic density and composition, and type of street and background concentration for calculating concentrations on the roadside (5 to 30 m from the main road). The results to date have had limits to validation because of the low number of measurements characteristic for traffic situations. The accuracy of results for benzo[a]pyrene using CAR calculations is therefore unknown. What is known for CAR is the following:

- In the determination of emission factors for traffic, performance and composition of the Netherlands car fleet, split into the built-up area, motorways and other roads, are taken into consideration. Using a component profile the emission factors for benzo[a]pyrene for light and heavy traffic are determined from the (total) emission factors for PAH. Finally, differentiation is introduced into the standard traffic limit profile for the built-up area.
- Traffic data are known for about 20 towns (out of 80) of more than 40,000 inhabitants. The relevant municipalities have supplied a traffic environment map (VMK), giving the necessary data for a calculation using the CAR model.
- Although the background concentrations for benzo[a]pyrene are not well known for the Netherlands, this background concentration is essential for establishing CAR parameters so as to calculate the regional background (i.e. for regions in the Netherlands) and the town's contribution. These parameters for benzo[a]pyrene are estimated using measurement data from the provinces of Noord-Holland and DCMR Rijnmond.
- There are too few measurement data on roadside locations to enable validation of the model results.

The stretch of road measured in kilometres exceeding the limit value can be derived after extrapolation to national scale from CAR model results and the VMK file.

Table 2 Total road length (km) in towns where exceedance of the limit value for benzo[a]pyrene (1 ng m^{-3}) occurs (RIVM, 1999).

Year	Road length
1990	190
1995	70
1997	50
1998	20

3.2 Monitoring

For the sake of simplicity this section will distinguish four different types of situations: background, situations near industrial sources, urban areas and other situations.

3.2.1 Background levels

The background level in this report is interpreted as being the level in situations outside the direct sphere of influence of point and surface sources in the Netherlands. The regional background level of PAH in the Netherlands can be estimated using the results of the measurements taken in De Zilk (North Sea coast), De Rijp (in the middle of Noord-Holland) and Vredepeel (southeast Netherlands). The average benzo[a]pyrene concentrations at these monitoring stations are about 0.05 ng m^{-3} to just above 0.10 ng m^{-3} (see also Table 3). The only long-range measurement series concerns measurements at De Rijp station (see Figure 3).

Table 3 Average level of a number of PAHs (ng m^{-3}) at background locations in the Netherlands around 1998-99. For specific information on time periods see Table 1.

Component	De Zilk (~1998)[1)]	De Rijp (1998)	Vredepeel (1998/99)
Benzo[a]pyrene	**0.06**	**0.11**	**0.08**
Benzo[b]fluoranthene	0.16	0.25	0.31
Benzo[ghi]perylene	0.15	0.18	0.21
Benzo[k]fluoranthene	0.06	0.11	0.11
Dibenzo[ah]anthracene	0.01	0.06	0.03
Indeno[123-cd]pyrene	0.09	0.14	0.23

[1)] Only a small part of 1998; see also Table 1.

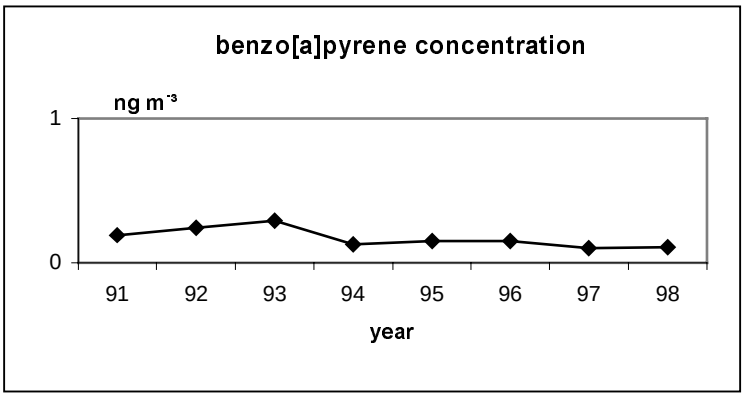

Figure 3 Yearly average benzo[a]pyrene concentrations at De Rijp monitoring station, 1991-1998.

3.2.2 Near industrial sources

In this section measurement results are presented which are explicitly meant to establish the situation in the immediate vicinity of industrial sources, namely, the measurements for the province of Zeeland at a cokes plant at Sluiskil in 1995/96[1], and of the province of Noord-Holland at the Hoogovens (Steelworks). In the last case, measurements were taken in Beverwijk by the RIVM as a supplement to the measurements taken for the province in 1998/99. The average of a number of PAHs observed at Sluiskil are noted in Table 4.

Table 4 Average levels of a number of PAHs (ng m^{-3}) at the Sluiskuil measuring station, 1995/96.

Component	1995	1996
Benzo[a]pyrene	**0.69**	**0.87**
Benzo[b]fluoranthene	1.8	1.5
Benzo[ghi]perylene	0.92	0.92
Benzo[k]fluoranthene	0.44	0.62
Dibenzo[ah]anthracene	0.79	0.50
Indeno[123-cd]pyrene	1.9	1.1

Ever since the 1980s the province of Noord-Holland has been monitoring near the Hoogovens (Steelworks) complex in IJmuiden/Beverwijk. This has resulted in an elaborate set of measurements for air quality in the vicinity of the Hoogovens. The measurements in Wijk aan Zee and, to a lesser extent, in IJmuiden give us a good idea of the environmental pollution caused by this PAH assumed source of substantial proportion. Table 5 shows the long-range averages for a number of PAHs, while Table 6 gives an impression of the situation in 1998/99. Figure 4 presents the yearly averages for benzo[a]pyrene concentrations over a number of years. A good interpretation is, however, made difficult as the yearly averages for Noord-Holland are calculated on the basis of 30-40 observations per calendar year. The presence or absence of a limited number of sharply increased levels in the data set has therefore a large influence on the average. In this way it is possible that the absence of these increased levels in the data set for 1993 will yield a much lower average than in the other years in which similar increased levels were observed.

Table 5 Long-range average levels of a number of PAHs (ng m^{-3}) at the Wijk aan Zee and IJmuiden stations.

	Wijk aan Zee	IJmuiden
Benzo[a]pyrene	**0.70**	**0.43**
Benzo[b]fluoranthene	1.6	1.5
Benzo[ghi]perylene	1.2	0.87
Benzo[k]fluoranthene	0.82	0.48
Dibenzo[ah]anthracene	0.14	0.09
Indeno[123-cd]pyrene	1.2	0.87

[1] This plant will, by the way, soon be shut down.

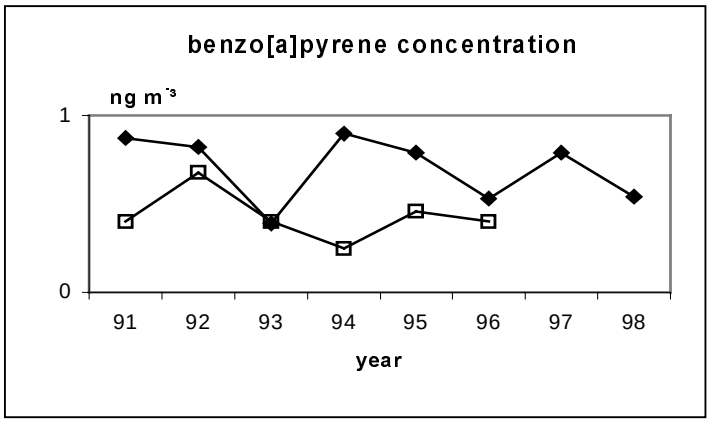

Figure 4 Yearly average for benzo[a]pyrene concentrations at the Wijk aan Zee (1991-1998, ◻) and IJmuiden stations (1991-1996, ■).

Table 6 Average level of a number of PAHs (ng m^{-3}) at stations near the Hoogovens, ca. 1998/99 (see Table 1 for exact information on time periods).

Component	Wijk aan Zee (1998)	Beverwijk (1998/99)
Benzo[a]pyrene	**0.54**	**0.11**
Benzo[b]fluoranthene	1.2	0.37
Benzo[ghi]perylene	1.1	0.30
Benzo[k]fluoranthene	0.48	0.12
Dibenzo[ah]anthracene	0.17	0.03
Indeno[123-cd]pyrene	1.0	0.30

3.2.3 Urban surroundings

Measurements for urban surroundings were taken by DCMR at two locations in Rotterdam and by RIVM at three locations in Utrecht. The Rotterdam-Vasteland locations can be categorised as city background, while the Rotterdam-Statenweg location nearby a busy through street, where measurements were carried out more than two years ago, is characteristic of a real street station. The difference in the influence of traffic here is clearly seen in the measurement results, as summarised in Table 7. Figure 5 shows the development of benzo[a]pyrene concentration measured at the Rotterdam stations over several years.

Table 7 Average PAH levels (ng m^{-3}) measured at the stations Rotterdam-Vasteland (1991/98) and Rotterdam-Statenweg (1994/95).

Component	Vasteland	Statenweg
Benzo[a]pyrene	**0.40**	**0.68**
Benzo[b]fluoranthene	0.53	0.79
Benzo[ghi]perylene	0.59	1.1
Benzo[k]fluoranthene	0.21	0.33
Dibenzo[ah]anthracene	0.22	0.95
Indeno[123-cd]pyrene	0.51	0.96

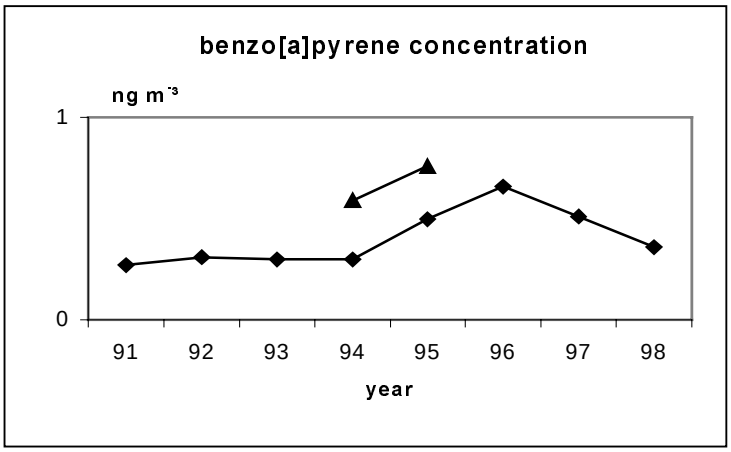

Figure 5 Benzo[a]pyrene at the Vasteland city station (1991/98,■) and the Statenweg road station (1994/95,▲).

Table 8 provides an overview of the recent measurements at the urban stations in Utrecht and Rotterdam.

Table 8 Average levels of a number of PAHs (ng m^{-3}) at urban stations, ca. 1998/99 (see Table 1 for exact information on time periods).

Component	Rotterdam Vasteland (1998)	Utrecht UB garden (1998/99)	Utrecht Erzeijstraat (1998/99)	Utrecht Wittevrouwen Straat (1998/99)
Benzo[a]pyrene	**0.36**	**0.13**	**0.25**	**0.35**
Benzo[b]fluoranthene	0.44	0.41	0.66	0.72
Benzo[ghi]perylene	0.51	0.36	0.88	0.91
Benzo[k]fluoranthene	0.14	0.15	0.24	0.30
Dibenzo[ah]anthracene	0.17	0.04	0.06	0.07
Indeno[123-cd]pyrene	0.25	0.33	0.57	0.61

The earlier measurements by RIVM (Van Velze, 1996) concern a city station in The Hague and a street station in Apeldoorn. The levels measured at The Hague station are of the same magnitude as at the Vasteland location. The results in Apeldoorn work out to a factor of 2 higher than the measured concentrations at the locations in Table 8.

3.2.4 Other situations

Besides the measurements presented in the previous sections, measurements have also been carried out in the vicinity of Schiphol. Seen from a monitoring-strategic perspective, the stations installed here do not belong to any of the previous categories. The situation in this area is described, in appropriate terms, as an area where the air is of urban quality (RIVM, 1999). Nevertheless, the Badhoevedorp station, and to lesser extent, the Hoofddorp station, also have more-or-less the character of traffic stations because of their proximity to (extremely) heavy traffic. Yearly average concentrations of a number of PAHs are presented in Table 9.

Table 9 Average levels of a number of PAHs (ng m^{-3}) for monitoring stations near Schiphol (1995/98).

	Badhoevedorp	Hoofddorp	Oude Meer
Benzo[a]pyrene	**0.29**	**0.25**	**0.27**
Benzo[b]fluoranthene	0.64	0.59	0.58
Benzo[ghi]perylene	0.58	0.47	0.57
Benzo[k]fluoranthene	0.48	0.24	0.23
Dibenzo[ah]anthracene	0.07	0.07	0.07
Indeno[123-cd]pyrene	0.45	0.39	0.43

4 Discussion

The limited number of PAH measurements taken in the Netherlands makes it difficult to give a reliable assessment of the air quality in this country. This limitation leads to a situation in which only indications can be given about the PAH levels. The sampling problems brought up earlier are responsible for the emphasis on less volatile PAHs, in particular, benzo[a]pyrene, in the following sections. It may also be noted that not only were measurements taken at few locations, but these were also limited in their time spans. This means that derived measurements, like yearly averages, are plagued by great uncertainties.

The RIVM monitoring in Utrecht at the three urban stations was at that time carried out simultaneously, allowing a direct intercomparison, since measurements relate to the same period. In Figure 6, which shows all the observations, benzo[a]pyrene behaviour at all stations is seen to keep tread with the time. This points to a large-scale dispersion pattern, with urban background contributions above the background levels (see the Utrecht UB garden and urban background station), and above these an extreme locally related contribution (see Utrecht-Erzeijstraat and Utrecht-Wittevrouwenstraat). Factors such as buildings and traffic intensity undoubtedly play a role here. The increased concentrations in weeks 8-10 resulted from the meteorological dispersion circumstances. The situation at the time was, in general, characterised by increased air pollution.

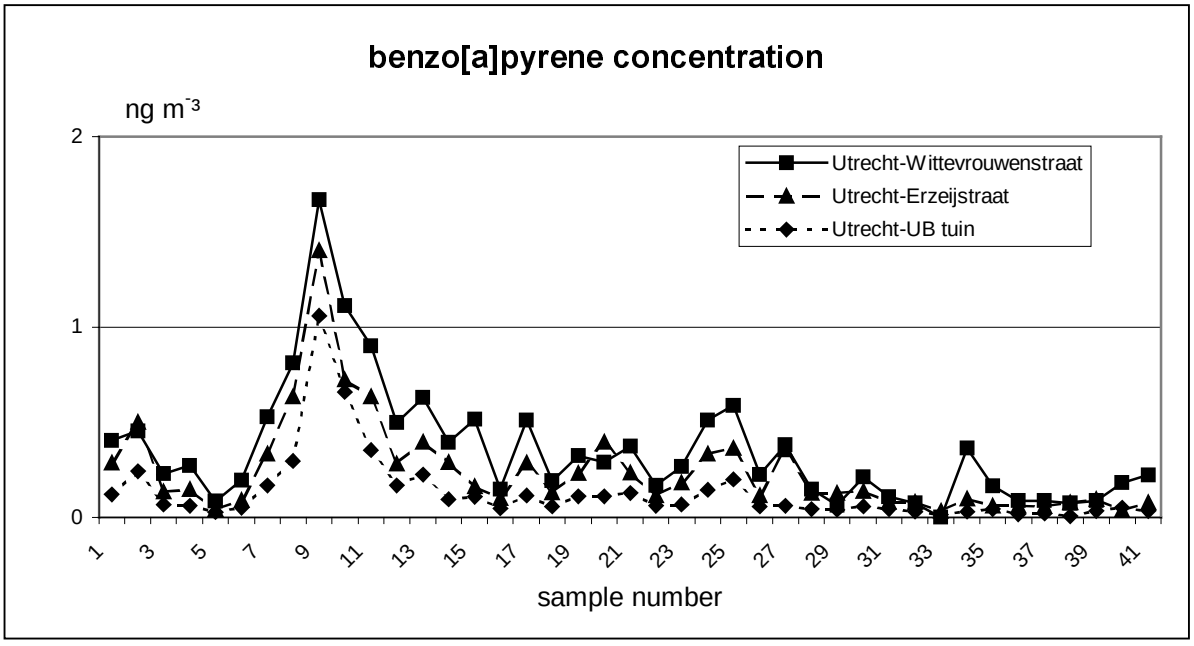

Figure 6 Weekly averages of benzo[a]pyrene at three urban stations in Utrecht, September 1998-June 1999.

The measurements at the urban stations show no exceedance of the limit value for benzo[a]pyrene. According to calculations, however, standards should be exceeded along a length of road of ca. 20 km (pointed out in Table 2). This discrepancy means that both the calculations and the measurements (still to be quantified) are plagued with uncertainties. Further research should lead to improvement in the quality of the statements made here.

A further remark can be made on the relative low (average) benzo[a]pyrene level of 0.11 ng m^{-3} measured in Beverwijk, where there is hardly ever mention of an increase in background level. On the other hand, the average level in Wijk aan Zee (ca. 0.5 ng m^{-3}) is comparably high. Although the level here is usually low, due to a limited number of extremely high levels observed, the yearly average has turned out high (see Figure 7). The model calculations making use of the most recent emission data (1996) for the Hoogovens would seem to confirm the measurement results. Besides the uncertainty in the emission data, there is more uncertainty present; this is because model calculations use emission data on PAH, like the 10 VROM PAH. Estimations of the benzo[a]pyrene concentrations resulting from the model calculations demand that PAH be recalculated to benzo[a]pyrene. The RIVM measurements in Beverwijk show benzo[a]pyrene to comprise about 0.7%[1] of the 10 VROM PAH. Using these recalculation factors in the model calculations, the resulting benzo[a]pyrene concentration is 0.15 ng m^{-3} for Beverwijk and 0.35 ng m^{-3} for Wijk aan Zee. The background level of 0.05 - 0.10 ng m^{-3} has to be added to each of these figures. The results of model calculations will be proven just as large as the measurement results.

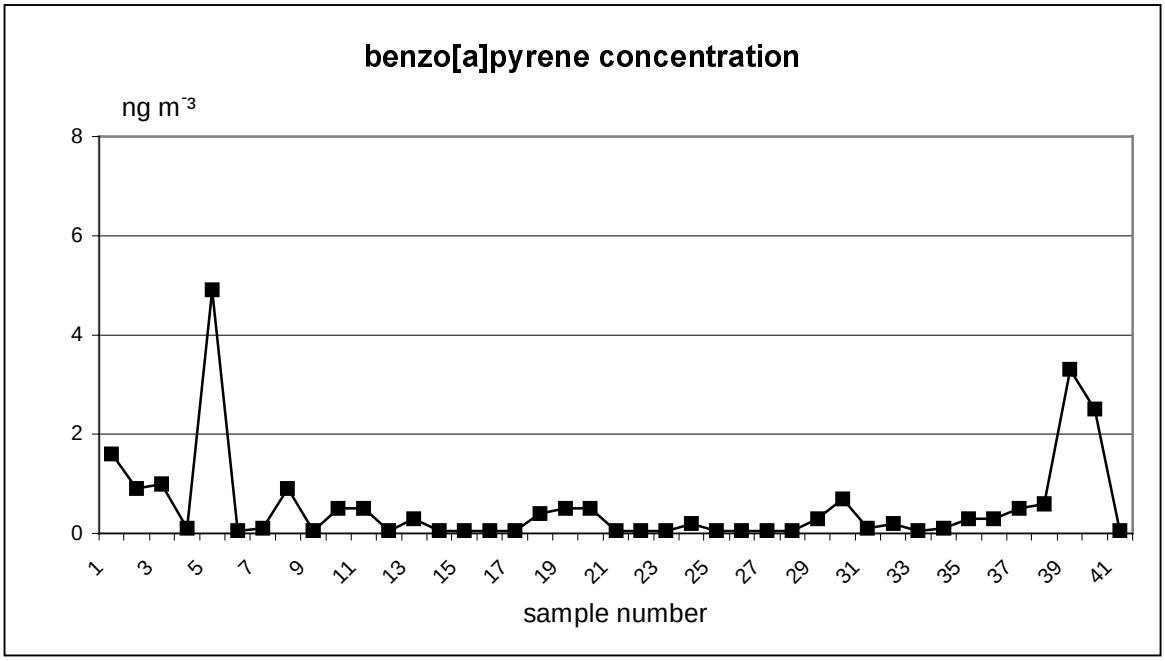

Figure 7 Benzo[a]pyrene concentrations (ng m^{-3}) from one sample per 8 days at Wijk aan Zee, 1998.

In the literature mention is made of some PAHs or PAH relationships being characteristic for certain contamination situations (Harrison *et al.*, 1996). Cokes manufacture is supposed to be recognised by its increased levels of anthracene, benzo[a]pyrene, benzo[ghi]perylene and fenantrene. In traffic, benzo[ghi]perylene and pyrene may be raised; a lot of diesel traffic will, furthermore, lead to increased levels of benzo[b]fluorathene and benzo[k]fluorathene. The ratio of indeno[123-cd] pyrene to benzo[ghi]perylene could be an indication of the traffic composition. A low ratio of indeno[123-cd]pyrene to benzo[b]pyrene points to a relatively large portion of traffic with petrol engines, whereas a high ratio points to a lot of traffic with diesel engines.

The results of the monitoring taking place in this investigation provide at first sight no clear confirmation of the above results, and an elaborate analysis of this aspect, for example, using factor analyses, falls outside the context of this project. In general, the concentrations of most of the PAHs can be said to rise in a situation which is potentially contaminable.

[1] This value does not appear to be constant; at urban stations the benzo[a]pyrene component is 0.8-1.0% and will rise even higher as the diesel component of traffic becomes larger.

5 Conclusions

The presence of PAH, and then especially the PAH indicator benzo[a]pyrene, in air in the Netherlands has been investigated in the project ‘Scanning and scouting air measurements'. 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. Statements on air quality for benzo[a]pyrene are largely based on results of measurements. Model calculations are particularly focused on potential problem situations caused by industrial sources.

The outcomes of the model calculations are uncertain. In the first place, PAH emission data are known for their uncertainties. In the second place, PAH has to be converted into benzo[a]pyrene, which introduces an extra uncertainty. Nevertheless, we can cautiously conclude that as far as industrial areas go, increased concentrations of benzo[a]pyrene may occur but only in the immediate vicinity of the Hoogovens.

The measurement data discussed in the report can be summarised into a general characterisation of a number of typical types of ‘areas’, as shown in Table 10. The figures presented may, however, show a large uncertainly margin, since these data are, generally speaking, based on limited data sets. Comparison with the current Dutch guide and limit values[1] shows us that the limit value is not exceeded; however, benzo[a]pyrene concentrations between the guide and limit values can be found at a number of stations, especially in streets where traffic is heavy.

The Wittevrouwenstraat in Utrecht was incorporated into this supplemental monitoring programme. This location is, within the context of the National Air Quality Monitoring Network, considered characteristic for high pollution caused by traffic. The limit value for benzo[a]pyrene was, however, not exceeded, which could mean that also in other areas where the traffic is heavy no exceedance of the limit value occurred.

Table 10 Average benzo[a]pyrene (ng m^{-3}) concentrations for a number of situations, based on monitoring.

Situation	Based on monitoring at:	Concentrations
Background	De Zilk, De Rijp, Vredepeel	0.05-0.15
Near industries[1)]	Hoogovens	0.2-0.5
City background	Rotterdam, Schiphol, Utrecht	0.2-0.4
Streets	Rotterdam, Utrecht	0.3-0.7

[1)] In a limited number of situations only.

[1] Limit value of 1 ng B[a]Pm^{-3} and guide value of 0.5 ng B[a] m^{-3}; both are yearly averages.

Acknowledgements

We gratefully acknowledge the experience and expertise provided by TNO-MEP, the Department of Environmental Quality and Analysis, the province of Noord-Holland and DCMR (advisory environmental service) Rijnmond in the inception phase of this project. Databases containing measurements carried out by Noord-Holland and Zeeland, as well as by DCMR, were of value in the reporting phase. The resulting measurements were made available by the aforementioned organisations. More information on a number of individual sources was supplied by Noord-Holland and Zeeland. Ruth de Wijs-Christensen is thanked for editorial assistance.

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Appendix A Mailing list

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38 Bibliotheek RIVM
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66-100 Reserve exemplaren

Appendix B Chemical and physical properties of a number of PAHs

Compound	Formula	Molecular mass	Vapour pressure 20° C (Pa)	Water solubility 20° C (mg l^{-1})	Log Kow	Toxicity Equivalent (TEF)
Fenantrene	$C_{14}H_{10}$	178	9.0×10^{-3}	1.3×10^{0}	4.46	0.001
Anthracene	$C_{14}H_{10}$	178	1.7×10^{-3}	7.0×10^{-2}	4.46	0.01
Fluoranthene	$C_{16}H_{10}$	202	7.9×10^{-5}	2.6×10^{-1}	5.33	0.001
Pyrene	$C_{16}H_{10}$	202	9.1×10^{-6}	1.4×10^{-1}	5.32	0.001
Benzo[a]anthracene	$C_{18}H_{12}$	228	6.6×10^{-8}	1.4×10^{-2}	5.61	0.1
Chrysene	$C_{18}H_{12}$	228	8.3×10^{-6}	2.0×10^{-3}	5.61	0.01
Benzo[b]fluoranthene	$C_{20}H_{12}$	252	6.6×10^{-6}	1.2×10^{-4}	6.57	0.1
Benzo[k]fluoranthene	$C_{20}H_{12}$	252	6.6×10^{-6}	5.5×10^{-4}	6.87	0.1
Benzo[a]pyrene	$C_{20}H_{12}$	252	6.6×10^{-6}	3.8×10^{-3}	6.04	1
Dibenzo(a.h)anthracene	$C_{22}H_{14}$	278	1.3×10^{-9}	5.0×10^{-4}	5.97	5
Benzo[ghi]perylene	$C_{22}H_{12}$	278	1.3×10^{-9}	2.6×10^{-4}	7.23	0.01
Indeno[123-cd]pyrene	$C_{22}H_{12}$	276	1.3×10^{-9}	6.2×10^{-2}	7.66	0.1

Appendix C Standard dispersion calculations

Standard dispersion matrices were 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 tonne 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, for example, limit values to see if further research was necessary. The advantage of this approach is that only a limited number of dispersion calculations have to be carried out. Figure C1 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 6941023 515 624 555 583 586 575 559 538 509
339 334 340 342 339 327 297 232 142 240 7911439 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 C1 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.

Table C1 gives the sources that were at first evaluated by standard dispersion matrices and for which high levels were concluded to be possible. This table also shows first-order estimates for the maximum concentration and the distance D of the maximum. Concentrations are expressed as the 10 VROM PAH (see also Appendix D); an indication of the order of magnitude of the corresponding benzo[a]pyrene concentrations can be obtained by multiplying the given numbers by 0.007. Based on the results from Table C1, it was concluded that only for the Hoogovens (Steelworks) do supplementary dispersion calculations on a more spatially detailed level have to be carried out.

Table C1 First-order estimates of maximum PAH concentrations based on data from the Emission Registration for the year 1995. Maximum benzo[a]pyrene concentrations can be estimated by multiplying C_{max} by 0.007.

Source	Location	C_{max} ($\mu g\ m^{-3}$)	D (in m)[1)]	Remark
Hoogovens	IJmuiden	25	<100	Several sources
Norit	Zaandam	12	<100	See note 2
Weweler	Apeldoorn	5	<100	
DAF Trucks	Eindhoven	4	<100	
Aluminium Chemie	Rotterdam	2	<100	
Pechiney	Vlissingen	2	<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.
[2)] On inquiry at the province of Noord-Holland, the responsible government authority, it appeared that emissions were primarily in the form of naphthalene and other volatile PAHs. According to recent information the actual emission is now lowered by a factor of 1000.

Models
The standard dispersion matrices, for which the results have been presented in Chapter 3, have been 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 D PAH compounds, series and monitoring programmes

Compound	Emission series			Monitoring programmes			
	Borneff	VROM -10	EPA-16	DCMR[1)]	Noord-Holland[2)]	RIVM	Zeeland
Acenaphthene			•	[•]	•		•
Acenaphthylene			•	[•]			•
Anthracene		•	•	•		•	•
Benzo[a]anthracene		•	•	•	•	•	•
Benzo[a]pyrene	•	•	•	•	•	•	•
Benzo[b]fluoranthene	•		•	•	•	•	•
Benzo[ghi]perylene	•	•	•	•	•	•	•
Benzo[k]fluoranthene	•	•	•	•	•	•	•
Chrysene		•	•	•	•	•	•
Corene					•		
Dibenzo[ah]anthracene			•	•	•	•	•
Fenanthrene		•	•	•		•	•
Fluoranthene	•	•	•	•		•	•
Fluorene			•			•	•
Indeno[1,2,3-cd]pyrene	•	•	•	•		•	
Indeno[gh]pyrene				[•]			•
Naphthalene		•	•	[•]			•
Pyrene			•	•	•	•	•

[1)] Compounds indicated with [•] have been measured to a limited extent.
[2)] The monitoring programme of OMEGAM is identical to that of the province of Noord-Holland.

Appendix E Sampling and analysis of PAHs

Measurements/RIVM

PAH sampling by RIVM takes place, to a large extent, according to the Dutch standard NVN 2798. Air with a flow of 1.6 $m^3 h^{-1}$ was sampled for a period of seven days; the total sampled volume was approximately 270 m^3. Glass fibre filters from Gelman, type A/E, were used. PUFF filters were made from commercially available polyurethane foam with a density of 35 kg m^{-3}. Before sampling, the PUFF was cleaned with dichloromethane and subsequently stored in plastic bags until mounting in the sampling equipment. After sampling, the filters were transported and stored under conditioned circumstances. The Laboratory of Organic Analytical Chemistry at RIVM carried out an analysis of PUFF and filters. The samples were extracted with Microwave-Assisted Solvent Extraction (MASE). PUFF and glass fibre filters were put in a special small container; an internal standard with 6-methylchrysene and D12-benzo[k]fluoranthene in acetone and more extraction liquid was added. Samples were extracted in a microwave oven at 115°C for 10 minutes. Next, a part of the extract was diluted with water (in a ratio 2:3), after which the samples were ready for analysis. Sometimes PAH concentrations in the PUFF extracts appeared to be so high that dilution was necessary in order to stay within the linear range of the detector. Analysis was carried out with an on-line SPE (Solid Phase Extraction) RP-HPLC (Reversed Phase High Pressure Liquid Chromatography) system and a fluorescence detector. Concentrations were calculated on an external standard mixture of 15 PAHs. Anthracene and benzo[a]anthracene results in PUFF were not reliable because of interferences in the PUFF. Acenaphthene in PUFF could not be analysed because of an interfering compound. Under naphthalene, fluorene and fenanthrene in PUFF, there were interferences from the HPLC system. However, correction is possible with the results of the analysed blanks. In the glass fibre filters there were a number of PAHs for which the results (naphthalene, acenaphthene, fluorene and fenanthrene) are not reliable due to the volatility of these PAHs. Field and laboratory blanks were taken on a regular basis.

Measurements/Noord-Holland

Sampling by the province of Noord-Holland was carried out with Sierra/Andersen PM10 model 1200 high volume samplers with a PM10 inlet. Whatman QM-A Quartz Microfibre Filters were used. The sampling period was one day; the sampled volume was 1625 m^3. No backup filter (or medium) was used for the volatile PAHs.

Measurements/OMEGAM

The procedures were almost completely identical to the procedures given under 'Measurements/Noord-Holland'. The one exception was the sampling time of 3.5 days and thus a sample volume of 5687 m^3.

Measurements/Zeeland

No information available.

Measurements/DCMR

The sampling flow was 2 $m^3 h^{-1}$. With a sampling time of one day the sampled volume is 48 m^3. The sampling tube meeting the Dutch NVN 2798 was placed in the open air. The tube was filled with PUFF as an absorbency material; on one side it was covered with a 10µm Teflon filter. Analysis of PUFF and the Teflon filter was carried out by IWACO with HPLC/fluorescence. The analysis results were randomly controlled by GC-MS/MS. SRM 1650 (originally 1649) was used as a reference material.

General remarks on the sampling

A summary of the most important characteristics of the different sampling methods is given in Table E1. Incomplete sampling of more volatile PAH can occur if only a filter (and no back-up medium) is used. The concentrations of a number of the PAHs reported by the province of Noord-Holland could therefore be too low (see also Baek *et al*., 1991, Harrison *et al*., 1996, Smith & Harrison, 1996).

The dispersion of PAH over filter and backup medium as a result of the RIVM measurements is shown in Table E2. These results are in accordance with the results of other experiments (Baek *et al*., 1991, Harrison *et al*., 1996, Smith & Harrison, 1996).
'Medium volume sampling' methods, which, for instance, were used by DCMR and RIVM, are very suitable for PAH sampling. Although these methods use a relatively low flow (and thereby low sampling speed), the (anthropogenic) PAHs are quantitatively sampled. Research has shown that most of the (anthropogenic) PAHs are bounded to particles with a diameter less than 4 µm (Van Vaeck & Van Cauwenberghe, 1985).

If sampling was carried out with a filter and a backup medium, the results reported here are given as the sum of what has been measured on the filter and on the backup medium.

Table E1 Characteristics of sampling methods by the different organisations/networks.

Organisation/network	Flow (m^3 h^{-1})	Period	Sampling	Remark
DCMR Milieudienst Rijnmond				
	2.0	24 hours	Teflon filter + PUFF	
	68	24 hours	Quartz fibre filter	
Province of Noord-Holland				PM10 inlet
Province of Zeeland	-[1)]	24 hours	-[1)]	
RIVM	1.6	7 days	Glass fibre filter + PUFF	
OMEGAM	68	3.5 days	Quartz fibre filter	PM10 inlet

[1)] Information not available

Table E2 Dispersion of PAH over filter and PUFF; results from the RIVM measurements.

Almost completely On the filter (90-100%)	To large extent on the filter (>50-90%)	To large extent on the PUFF (>50-90%)	Almost completely on the PUFF (90-100%)
Benzo[a]pyrene	Chrysene	Benzo[a]anthracene	Anthracene
Benzo[b]fluoranthene			Fenanthrene
Benzo[ghi]perylene			Fluorene
Benzo[k]fluoranthene			Naphthalene
Dibenzo[ah]anthracene			
Indeno[123-cd]pyrene			

Appendix F Measurement results

Table F1 Results of PAH measurements (ng m^{-3}) at Sluiskil taken by the province of Zeeland; 24-hour sampling, every three days.

Component	1991	1992	1993	1994	1995	1996	1997	1998
Number of measurements	-	-	-	-	*111*	*107*	-	-
Acenaphthene	.	.	.	.	4.7	7.2	.	.
Acenaphthylene	.	.	.	.	20	12	.	.
Anthracene	.	.	.	.	2.3	2.5	.	.
Benzo[a]anthracene	.	.	.	.	1.5	1.7	.	.
Benzo[a]pyrene	.	.	.	.	**0.69**	**0.87**	.	.
Benzo[b]fluoranthene	.	.	.	.	1.8	1.5	.	.
Benzo[ghi]perylene	.	.	.	.	0.92	0.92	.	.
Benzo[k]fluoranthene	.	.	.	.	0.44	0.62	.	.
Chrysene	.	.	.	.	1.79	1.8	.	.
Dibenzo[a.h]anthracene	.	.	.	.	0.79	0.50	.	.
Fenanthrene	.	.	.	.	22	19	.	.
Fluoranthene	.	.	.	.	8.7	7.6	.	.
Fluorene	.	.	.	.	15	12	.	.
Indeno[123-cd]pyrene	.	.	.	.	1.9	1.1	.	.
Naphthalene	.	.	.	.	23	26	.	.
Pyrene	.	.	.	.	4.9	4.6	.	.

Table F2 Results of PAH measurements (ng m^{-3}) at Rotterdam-Vasteland by DCMR; 24-hour sampling, every eight days. Concentrations are the sums of filter and backup medium results.

Component	1991	1992	1993	1994	1995	1996	1997	1998
Number of measurements	*46*	*42*	*24*	*48*	*55* /	*42*	*28*	*56*
Acenaphthene	.	.	.	.	2,4	5,1	2,8	2,4
Acenaphthylene	.	.	.	.	7.8	9.7	3.8	6.2
Anthracene	1.8	2.0	1.2	1.9	1.9	3.9	2.2	1.2
Benzo[a]anthracene	0.24	0.51	0.41	0.45	0.47	0.64	0.37	0.22
Benzo[a]pyrene	**0.27**	**0.31**	**0.30**	**0.30**	**0.50**	**0.66**	**0.14**	**0.36**
Benzo[b]fluoranthene	0.38	0.22	0.60	0.47	0.66	0.78	0.65	0.44
Benzo[ghi]perylene	0.48	0.28	0.54	0.50	0.69	0.87	0.86	0.51
Benzo[k]fluoranthene	0.19	0.11	0.27	0.21	0.27	0.33	0.81	0.14
Chrysene	0.61	1.4	1.0	0.88	0.75	1.1	0.38	0.46
Dibenzo[a.h]anthracene	0.55	0.07	0.07	0.11	0.24	0.34	0.19	0.17
Fenanthrene	29	38	15	22	24	33	38	19
Fluoranthene	15	7.5	6.6	7.2	6.2	11	8.3	5.2
Fluorene	.	.	.	.	9.4	20	11	8.7
Indeno[123-cd]pyrene	0.39	0.13	0.39	0.25	0.88	0.97	0.81	0.25
Naphthalene	.	.	.	.	11	22	14	14
Pyrene	4.1	3.1	3.2	4.9	4.7	6.3	5.7	3.5

Table F3 Results of PAH measurements (ng m^{-3}) at Rotterdam-Statenweg taken by DCMR; 24-hour sampling, every eight days. Concentrations are the sums of filter and backup medium results.

Component	1991	1992	1993	1994	1995	1996	1997	1998
Number of measurements	-	-	-	*44*	*59*	-	-	-
Acenaphthene	.	.	.	.	.	.	.	.
Acenaphthylene	.	.	.	.	.	.	.	.
Anthracene	.	.	.	4.1	5.1	.	.	.
Benzo[a]anthracene	.	.	.	0.79	0.91	.	.	.
Benzo[a]pyrene	**.**	**.**	**.**	**0.59**	**0.76**	**.**	**.**	**.**
Benzo[b]fluoranthene	.	.	.	0.69	0.89	.	.	.
Benzo[ghi]perylene	.	.	.	0.86	1.4	.	.	.
Benzo[k]fluoranthene	.	.	.	0.32	0.34	.	.	.
Chrysene	.	.	.	1.3	1.2	.	.	.
Dibenzo[a.h]anthracene	.	.	.	0.11	0.31	.	.	.
Fenanthrene	.	.	.	46	51	.	.	.
Fluoranthene	.	.	.	15	14	.	.	.
Fluorene	.	.	.	.	.	.	.	.
Indeno[123-cd]pyrene	.	.	.	0.40	1.5	.	.	.
Naphthalene	.	.	.	.	.	.	.	.
Pyrene	.	.	.	10	11	.	.	.

Table F4 Results of PAH measurements (ng m^{-3}) at Wijk aan Zee[1)] by the province of Noord-Holland; 24-hour sampling, every six days.

Component	1991	1992	1993	1994	1995	1996	1997	1998
Number of measurements	*40*	*31*	*39*	*39*	*37*	*36*	*40*	*41*
Acenaphthene	.	.	.	.	.	.	.	.
Acenaphthylene	.	.	.	.	.	.	.	.
Anthracene	.	.	.	.	.	.	.	.
Benzo[a]anthracene	0.50	0.62	0.33	0.82	0.50	0.36	0.60	0.33
Benzo[a]pyrene	**0.87**	**0.82**	**0.39**	**0.90**	**0.79**	**0.53**	**0.79**	**0.54**
Benzo[b]fluoranthene	2.1	1.5	1.1	3.0	1.4	1.2	1.4	1.2
Benzo[ghi]perylene	1.2	1.1	1.0	2.1	0.99	0.88	0.94	1.1
Benzo[k]fluoranthene	0.88	0.60	0.43	1.4	0.70	0.54	0.63	0.48
Chrysene	0.90	0.96	0.62	1.5	0.71	0.70	0.90	0.40
Dibenzo[a.h]anthracene	0.15	0.12	0.13	0.22	0.13	0.10	0.13	0.17
Fenanthrene	.	.	.	.	.	.	.	.
Fluoranthene	.	.	.	.	.	.	.	.
Fluorene	0.54	0.55	0.42	1.6	0.72	0.85	0.72	0.28
Indeno[123-cd]pyrene	0.97	1.0	0.98	2.6	1.1	0.92	0.86	1.0
Naphthalene	.	.	.	.	.	.	.	.
Pyrene	0.53	0.54	0.36	0.54	0.48	0.43	0.43	0.28

[1)] 1991 through 1995 for Wijk aan Zee-Verlengde Voorstraat; from 1996 onwards for Wijk aan Zee-Banjaert

Table F5 Results of PAH measurements (ng m^{-3}) at IJmuiden by the province of Noord-Holland; 24-hour sampling, every six days.

Component	1991	1992	1993	1994	1995	1996	1997	1998
Number of measurements	*40*	*38*	*37*	*38*	*37*	*36*	.	.
Acenaphthene	.	.	.	.	.	.	.	.
Acenaphthylene	.	.	.	.	.	.	.	.
Anthracene	.	.	.	.	.	.	.	.
Benzo[a]anthracene	0.44	0.57	0.38	0.38	0.35	0.37	.	.
Benzo[a]pyrene	**0.40**	**0.68**	**0.40**	**0.25**	**0.46**	**0.40**	.	.
Benzo[b]fluoranthene	1.2	1.3	1.1	1.2	1.1	1.2	.	.
Benzo[ghi]perylene	0.72	0.89	0.91	0.99	0.84	0.96	.	.
Benzo[k]fluoranthene	0.43	0.52	0.40	0.52	0.48	0.51	.	.
Chrysene	0.81	0.99	0.71	0.88	0.63	0.79	.	.
Dibenzo[a.h]anthracene	0.07	0.10	0.11	0.09	0.10	0.08	.	.
Fenanthrene	.	.	.	.	.	.	.	.
Fluoranthene	.	.	.	.	.	.	.	.
Fluorene	0.55	0.73	0.52	0.60	0.74	0.72	.	.
Indeno[123-cd]pyrene	0.56	0.83	0.89	1.1	0.88	0.98	.	.
Naphthalene	.	.	.	.	.	.	.	.
Pyrene	0.54	0.72	0.45	0.42	0.47	0.42	.	.

Table F6 Results of PAH measurements (ng m^{-3}) at De Rijp by the province of Noord-Holland; 24-hour sampling, every six days.

Component	1991	1992	1993	1994	1995	1996	1997	1998
Number of measurements	*21*	*14*	*20*	*19*	*29*	*30*	*38*	*39*
Acenaphthene	.	.	.	.	.	.	.	.
Acenaphthylene	.	.	.	.	.	.	.	.
Anthracene	.	.	.	.	.	.	.	.
Benzo[a]anthracene	0.21	0.19	0.26	0.19	0.12	0.17	0.19	0.11
Benzo[a]pyrene	**0.19**	**0.24**	**0.29**	**0.13**	**0.15**	**0.15**	**0.10**	**0.11**
Benzo[b]fluoranthene	0.56	0.58	0.84	0.75	0.36	0.47	0.38	0.25
Benzo[ghi]perylene	0.30	0.42	0.68	0.58	0.27	0.32	0.32	0.18
Benzo[k]fluoranthene	0.20	0.20	0.30	0.33	0.15	0.20	0.17	0.11
Chrysene	0.45	0.40	0.59	0.50	0.21	0.37	0.24	0.15
Dibenzo[a.h]anthracene	0.06	0.06	0.09	0.07	0.06	0.06	0.06	0.06
Fenanthrene	.	.	.	.	.	.	.	.
Fluoranthene	.	.	.	.	.	.	.	.
Fluorene	0.48	0.34	0.43	0.34	0.21	0.36	0.32	0.19
Indeno[123-cd]pyrene	0.27	0.35	0.70	0.64	0.27	0.33	0.24	0.14
Naphthalene	.	.	.	.	.	.	.	.
Pyrene	0.43	0.29	0.35	0.26	0.14	0.22	0.22	0.12

Table F7 Results of PAH measurements (ng m^{-3}) at Badhoevedorp taken by the province of Noord-Holland. 24- hour sampling, every six days. (1995: every three days).

Component	1991	1992	1993	1994	1995	1996	1997	1998
Number of measurements	.	.	.	.	*107*	*35*	*39*	*40*
Acenaphthene	.	.	.	.	.	.	.	.
Acenaphtylene	.	.	.	.	.	.	.	.
Anthracene	.	.	.	.	.	.	.	.
Benzo[a]anthracene	.	.	.	.	0.26	0.31	0.30	0.24
Benzo[a]pyrene	**.**	**.**	**.**	**.**	**0.29**	**0.37**	**0.31**	**0.20**
Benzo[b]fluoranthene	.	.	.	.	0.69	0.84	0.55	0.49
Benzo[ghi]perylene	.	.	.	.	0.60	0.73	0.55	0.39
Benzo[k]fluoranthene	.	.	.	.	0.29	0.35	0.23	0.20
Chrysene	.	.	.	.	0.49	0.64	0.42	0.36
Dibenzo[a.h]anthracene	.	.	.	.	0.06	0.07	0.08	0.06
Fenanthrene	.	.	.	.	.	.	.	.
Fluoranthene	.	.	.	.	.	.	.	.
Fluorene	.	.	.	.	0.39	0.57	0.37	0.28
Indeno[123-cd]pyrene	.	.	.	.	0.52	0.65	0.38	0.23
Naphthalene	.	.	.	.				.
Pyrene	.	.	.	.	0.25	0.35	0.26	0.18

Table F8 Results of PAH measurements (ng m^{-3}) at Oude Meer taken by the province of Noord-Holland. 24- hour sampling, every six days. (1995: every three days).

Component	1991	1992	1993	1994	1995	1996	1997	1998
Number of measurements	.	.	.	.	*102*	*33*	*30*	*40*
Acenaphthene	.	.	.	.	.	.	.	.
Acenaphtylene	.	.	.	.	.	.	.	.
Anthracene	.	.	.	.	.	.	.	.
Benzo[a]anthracene	.	.	.	.	0.20	0.27	0.21	0.21
Benzo[a]pyrene	**.**	**.**	**.**	**.**	**0.26**	**0.30**	**0.19**	**0.24**
Benzo[b]fluoranthene	.	.	.	.	0.60	0.82	0.36	0.56
Benzo[ghi]perylene	.	.	.	.	0.48	0.66	0.33	0.42
Benzo[k]fluoranthene	.	.	.	.	0.24	0.33	0.16	0.21
Chrysene	.	.	.	.	0.38	0.65	0.25	0.39
Dibenzo[a.h]anthracene	.	.	.	.	0.06	0.07	0.07	0.06
Fenanthrene	.	.	.	.	.	.	.	.
Fluoranthene	.	.	.	.	.	.	.	.
Fluorene	.	.	.	.	0.30	0.61	0.28	0.31
Indeno[123-cd]pyrene	.	.	.	.	0.45	0.65	0.19	0.26
Naphthalene	.	.	.	.				.
Pyrene	.	.	.	.	0.19	0.37	0.20	0.23

Table F10 Results of PAH measurements (ng m^{-3}) at Hoofddorp taken by the province of Noord-Holland. 24- hour sampling, every six days (1994 and 1995: every three days).

Component	1991	1992	1993	1994	1995	1996	1997	1998
Number of measurements	.	.	.	*104*	*108*	*36*	*36*	*40*
Acenaphthene	.	.	.	.	.	.	.	.
Acenaphtylene	.	.	.	.	.	.	.	.
Anthracene	.	.	.	.	.	.	.	.
Benzo[a]anthracene	.	.	.	0.23	0.20	0.29	0.23	0.18
Benzo[a]pyrene	**.**	**.**	**.**	**0.17**	**0.29**	**0.37**	**0.25**	**0.16**
Benzo[b]fluoranthene	.	.	.	0.66	0.61	0.88	0.45	0.37
Benzo[ghi]perylene	.	.	.	0.70	0.62	0.83	0.50	0.29
Benzo[k]fluoranthene	.	.	.	0.29	0.24	0.36	0.18	0.15
Chrysene	.	.	.	0.45	0.40	0.64	0.33	0.26
Dibenzo[a.h]anthracene	.	.	.	0.06	0.06	0.07	0.07	0.06
Fenanthrene	.	.	.	.	.	.	.	.
Fluoranthene	.	.	.	.	.	.	.	.
Fluorene	.	.	.	0.34	0.32	0.57	0.32	0.19
Indeno[123-cd]pyrene	.	.	.	0.61	0.50	0.71	0.32	0.19
Naphthalene	.	.	.	.	.	.	.	.
Pyrene	.	.	.	0.24	0.20	0.34	0.23	0.15

Table F11 Results of PAH measurements (ng m^{-3}) at De Zilk taken by OMEGAM. Continuous sampling with a sampling period of 3.5 days.

Component	Concentration
Number of measurements	*19*
Acenaphthene	.
Acenaphtylene	.
Anthracene	.
Benzo[a]anthracene	0.04
Benzo[a]pyrene	**0.06**
Benzo[b]fluoranthene	0.16
Benzo[ghi]perylene	0.15
Benzo[k]fluoranthene	0.06
Chrysene	0.06
Dibenzo[a.h]anthracene	0.01
Fenanthrene	.
Fluoranthene	0.08
Fluorene	.
Indeno[123-cd]pyrene	0.09
Naphthalene	.
Pyrene	0.05

Table F12 Results of PAH measurements (ng m^{-3}) taken by RIVM. Average values based upon continuous sampling with a sampling period of 7 days in the period September 1998 through June 1999. Concentrations are the sums of filter and backup medium results.

Component	Utrecht 637 Traffic	Utrecht 639 Traffic	Utrecht 640 City background	Beverwijk 555 Source-directed	Vredepeel 131 Rural background
Acenaphthene	.	.	.	.	.
Acenaphtylene	.	.	.	.	.
Anthracene	4.4	3.2	1.1	1.6	0.68
Benzo[a]anthracene	1.5	1.5	0.90	1.1	1.0
Benzo[a]pyrene	**0.35**	**0.25**	**0.13**	**0.11**	**0.08**
Benzo[b]fluoranthene	0.72	0.66	0.42	0.37	0.31
Benzo[ghi]perylene	0.91	0.88	0.36	0.30	0.23
Benzo[k]fluoranthene	0.27	0.24	0.15	0.12	0.11
Chrysene	0.89	0.82	0.53	0.85	0.45
Dibenzo[a.h]anthracene	0.07	0.06	0.04	0.03	0.03
Fenanthrene	.	.	.	.	.
Fluoranthene	11	8.8	4.9	6.2	3.5
Fluorene	.	.	.	.	.
Indeno[123-cd]pyrene	0.60	0.57	0.33	0.30	0.23
Naphthalene	.	.	.	.	.
Pyrene	7.2	6.1	2.5	3.0	1.5

Table F13 Results of PAH measurements (ng m^{-3}) taken by RIVM (Van Velze, 1996). Average values based upon 24-hour samples, February through April 1992.

Compound	Apeldoorn 728 Traffic station	Den Haag 404 City background	Westmaas 437 Regional background
Number of measurements	*9*	*9*	*9*
Acenaphthene	.	.	.
Acenaphtylene	.	.	.
Anthracene	5.4	2.3	0.72
Benzo[a]anthracene	1.3	0.44	0.28
Benzo[a]pyrene	**0.91**	**0.31**	**0.27**
Benzo[b]fluoranthene	.	.	.
Benzo[ghi]perylene	1.9	0.68	0.45
Benzo[k]fluoranthene	0.70	0.31	0.27
Chrysene	3.1	1.4	0.94
Dibenzo[a.h]anthracene	0.58	0.07	0.12
Fenanthrene	39	22	17
Fluoranthene	17	8.4	5.4
Fluorene	.	.	.
Indeno[123-cd]pyrene	1.1	0.44	0.33
Naphthalene	.	.	.
Pyrene	15	6.9	