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# Long-term measurements of γ-HCH in precipitation in the Netherlands

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# Abstract

The results of 20 years of measuring γ-HCH in precipitation in the Netherlands are presented here. Analysis has revealed a systematic seasonal behaviour, with enhanced levels of γ-HCH in precipitation from April through June. Evidence was found for a statistically significant change in concentrations during the last 20 years. Measurement results from the last three years (1999-2001) showed a clear downward trend, suggesting a decline in lindane emissions. An evaluation of measurement results from northwest Europe confirms the large-scale distribution of γ-HCH in precipitation in northwest Europe. Given the large uncertainties in emission estimates and in modelling of atmospheric transport and deposition, measurements are concluded to form a better tool for monitoring the changes in environmental quality and emissions.

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# Samenvatting

De resultaten van 20 jaar metingen γ-HCH in neerslag in Nederland worden gepresenteerd. Analyse van de data toont aan dat er sprake is van een systematisch seizoensgedrag met verhoogde niveaus van γ-HCH in neerslag in de periode van april tot en met juni. Er is een statistisch significante verandering in de concentraties in de afgelopen 20 jaar gevonden. Een evaluatie van meetresultaten uit noordwest Europa bevestigt de grootschalige verspreiding van γ–HCH in neerslag in noordwest Europa. Op basis van de grote onzekerheden in emissieschattingen en in het modelleren van het atmosferisch transport en depositie wordt geconcludeerd dat metingen een beter instrument zijn om veranderingen in de milieukwaliteit en in de emissies te monitoren.

# Summary

The results of 20 years of measuring γ–HCH in precipitation in the Netherlands are presented here. Analysis has revealed a systematic seasonal behaviour, with enhanced levels of γ–HCH in precipitation from April through June. A statistically significant change in concentrations during the last 20 years was found. An evaluation of measurement results from north-west Europe confirms the large-scale distribution of γ–HCH in precipitation in that region. Long time series of γ–HCH measurements are a useful tool in evaluating its emission trend. Given the large uncertainties in emission estimates and in modelling of atmospheric transport and deposition up to now, measurements are considered to be a better tool for monitoring the changes in environmental levels of γ–HCH.

# 1. Introduction

HCH (hexachlorocyclohexane) has been used worldwide as a pesticide for almost 60 years (Li, 1999). Technical HCH is generally a mixture of five isomers in which α-HCH is the dominant one. However, γ-hexachlorocyclohexane (γ–HCH) is the isomer with the highest insecticidal activity. Lindane is composed of 90% (or more) γ-HCH [1]. This isomer shows several serious disadvantages, the most prominent being its persistence, its tendency to bioaccumulate and its toxicity. Due to these qualities γ–HCH is considered a Persistent Organic Pollutant (POP). Lindane is banned or severely restricted in many countries (Breivik *et al*., 1999) and forms part of the UN- ECE POPs protocol aiming to diminish use and emission of POP. Moreover, the atmospheric chemical stability of γ–HCH and its characteristic gaseous state in the atmosphere (Van Jaarsveld *et al*., 1997) contribute to the long-range transport of γ–HCH. Estimates of the atmospheric residence time of γ–HCH range from 5 to 100 days (*e.g.* Atkinson *et al*., 1992; Brubaker and Hites, 1998; Van Pul *et al*., 1998) corresponding to travel distances of thousands of kilometres or more. Research has shown that even in remote areas, *e.g.* in high mountain lakes and in the Arctic and Antarctica, substantial levels of γ–HCH occur in the environment as well as in mammals (Carrera *et al*., 2002; Oehme *et al*., 1996; Kallenborn *et al*., 1998). In laboratory studies γ–HCH has shown to lead to detrimental effects including endocrine-disrupting effects (Ulrich *et al*., 2000), hepatic changes and decreased survival (WHO, 1991).

The removal pathways of γ–HCH from the atmosphere are conversion by the hydroxyl radical and by wet and dry deposition. Since γ–HCH is relatively soluble in water, wet deposition is the dominant removal pathway above land surfaces, whereas dry deposition is more efficient above sea surfaces (Van Jaarsveld *et al*., 1997).

Long-term information on the environmental γ-HCH levels on the basis of systematic measurements is scarce, and so hampers an adequate evaluation of the environmental levels over longer periods of time. Indeed, γ-HCH in air and precipitation have been measured on a limited number of sites of the pan-European EMEP network, but only since 1992 (Berg *et al.,* 2001). Although considerable progress in the field of atmospheric modelling of γ-HCH has been achieved, the uncertainty in the modelling results is still large. Major obstacles are large uncertainties in estimates of γ-HCH emission data, the lack of long-term records of γ-HCH measurements in the environment –the results from the Integrated Atmopsheric Deposition Network in North America (see *e.g*. Hillary *et al.,* 1997) and the Arctic Monitoring and

Assessment programma (AMAP, 2000) being positive exceptions-, the uncertainty in deposition velocities and uncertainties in exchange rates with soil and water surfaces (Jacobs and Van Pul, 1996; Holoubek *et al.,* 2000; Dubus *et al.*, 2000). Evaluation of long-term measurement results will a) support a more reliable validation of modelling results, and with this, a more accurate description of γ-HCH environmental levels and can be b) used to evaluate the effects of the policy measures. Here, long-term measurements of concentrations of γ-HCH in precipitation in the Netherlands will be discussed and results placed in the context of measurement results from elsewhere. They will also be used to make an independent estimate of the emission history and will be compared with predictions from modelling efforts.

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[1] For the sake of clarity, only the name 'γ–HCH' will be used in this paper

# 2. Methods

Measurements of γ-HCH in precipitation started in the Netherlands in 1980. Measurements have been carried out since then on a monthly (or four weekly) basis in the Netherlands Precipitation Chemistry Network within the framework of the Netherlands Air Quality Monitoring Network (Van Elzakker, 2001). Sampling started at the measurement site of De Bilt on January 1st, 1980. Later on, more stations became operational for shorter or longer periods of time (see *Table 1* for an overview and *Figure 1*). There are, to the best of our knowledge, no local sources of lindane, and thus no γ-HCH emissions near the monitoring sites, nor have there been any in the past. All the sites were at the grounds of public organisations, like meteo-stations or water-collection areas, thereby guaranteeing the support of local personnel. Precipitation was sampled with a bulk sampler. The funnel of the sampler was made of Pyrex glass with a sampling surface of 500 $cm^2$. The sampling height was 1.50 meter above the surface. Precipitation was sampled in a 5-litre Pyrex bottle, protected from light. From 1980 till 1985, the sample bottle was placed in a hole in the ground and thus protected against light and kept relatively cool. As of 1985 the sample bottle was placed in a white-painted PVC housing and thus protected from light. The white painting prevents high temperatures inside the housing. Local personnel checked the sampling equipment every day and, if necessary, changed sample bottles. The length of the sampling period was one month (until 1991) or four weeks (from 1991 onwards). Each sampling period started with a thorough cleaning of the funnel with copious amounts of distilled water. At the end of the period the sampling bottles were transported to the RIVM's Organic-Analytical Laboratory in Bilthoven, where the analysis was carried out. In the laboratory samples were kept in a refrigerator until extraction. Extractions of the precipitation samples with hexane were carried out within two weeks after arrival of the samples at the laboratory. Drying the extract with anhydrous sodium sulphate followed extraction. Analysis was done on α-HCH (until 1990) and γ-HCH. From 1980 till 1993 analysis was done with capillary gas chromatography with an electron capture detector. From 1993 until 2000, samples in which γ-HCH was positively identified, were re-analysed using capillary gas chromatography with mass spectrometric confirmation and quantification. Since 2000, all samples have been directly analysed with capillary gas chromatography with mass spectrometric quantification. The limit of detection was 0.01 μg/l for all methods. Quality control procedures in the laboratory included regular recovery experiments at 0.01 μg/l and 0.10 μg/l levels, done, in general four times a year. Recovery experiments were done by spiking blank rainwater prior to the extraction procedure. Recoveries were 96% on average, ranging from 90 to 125%. Results presented here have not been corrected for these recoveries.

*Table 1 Measurement sites for γ–HCH in precipitation in the Netherlands, 1980-2001*

| Station name | Co-ordinates | From: | To: |
|---|---|---|---|
| De Bilt | 52°06'03"N 05°10'40"E | 1-1980 | 1-1999 |
| Vlissingen | 51°26'33"N 03°35'47"E | 1-1985 | 1-1988 |
| Witteveen | 52°48'49"N 06°40'11"E | 1-1985 | 1-1988 |
| Leiduin | 52°20'44"N 04°35'52"E | 1-1988 | 6-1993 |
| Rotterdam | 51°57'14"N 04°26'36"E | 1-1988 | 1-1992 |
| De Zilk | 52°17'53"N 04°30'37"E | 4-1994 | 1-2002 |

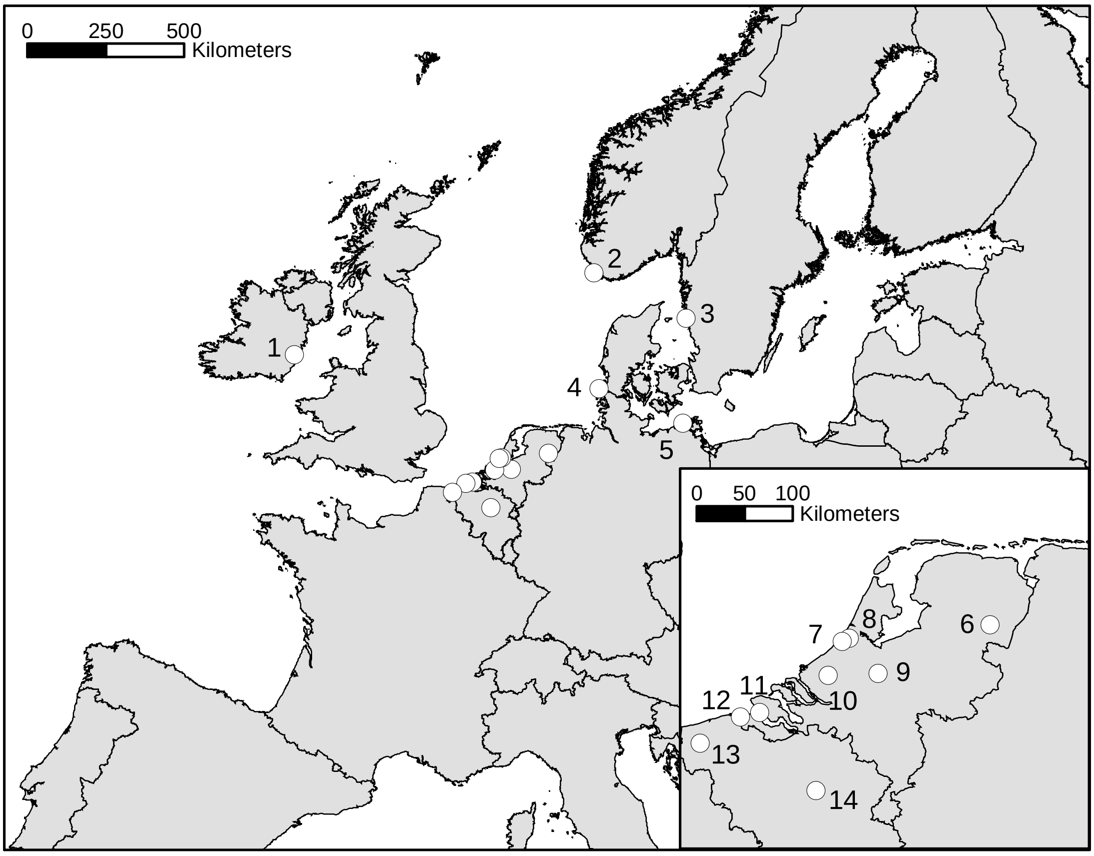

*Figure 1 Map showing measurement sites for γ–HCH in precipitation in Europe and the Netherlands, 1980-2001.1: Turlough Hill; 2: Lista; 3: Rörvik; 4: Westerland; 5: Zingst; 6: Witteveen; 7: De Zilk; 8: Leiduin; 9: De Bilt; 10: Rotterdam; 11: Vlissingen; 12: Knokke; 13: Blankaart; 14:Kessel (Stórhöfdi, Iceland, not shown).*

# 3. Results

In the 1980-2001 period 487 samples were taken, of which 13% showed a concentration below the limit of detection of the analytical method (0.01 µg/l). For nine samples, the sample volume was too low to perform an analysis, *ie* a volume of 100 ml or less corresponding to a precipitation amount of 12 mm or less. Thirteen samples were lost, mainly in wintertime due to the sample bottle freezing and subsequent cracking of the bottle. Summarised measurement of γ-HCH in precipitation results are shown in *Table 2*. Average values are here presented as 50th percentile (=median) values, and not as arithmetic or volume-weighted averages, because of the large number of relatively low concentrations.

*Table 2 Characteristics of γ–HCH in precipitation in the Netherlands, 1980-2001*

| Station | From: | To: | Number of samples | Of which with C< dl [1] | Range of peak values [2] | 50 percentile |
|---|---|---|---|---|---|---|
| | | | | | µg/l | µg/l |
| De Bilt | 1-1980 | 1-1999 | 203 | 20 | 0.07-2.00 | 0.02 |
| Vlissingen | 1-1985 | 1-1988 | 34 | 1 | 0.16-2.80 | 0.05 |
| Witteveen | 1-1985 | 1-1988 | 34 | 2 | 0.08-1.00 | 0.04 |
| Leiduin | 1-1988 | 6-1993 | 70 | 7 | 0.07-0.68 | 0.02 |
| Rotterdam | 1-1988 | 1-1992 | 50 | 5 | 0.10-0.68 | 0.02 |
| De Zilk | 4-1994 | 1-2002 | 96 | 29 | 0.02-0.68 | 0.01 |

[1] dl: detection limit (0.01 µg γ-HCH/l)
[2] The highest concentration value has been taken from each year.

The measurement sites of Leiduin (1988-1993) and De Zilk (from 1994 onwards) are fairly close to each other, approximately eight kilometres in a straight line. Thus, we could consider the data from these two stations as coming from the same underlying distribution.

Some periods allow a comparison of results from different locations: 1985-1987 (3 stations), 1988-1991 (3 stations) and 1994-1998 (2 stations). Measurement results from stations at a relatively close distance (Leiduin vs. Rotterdam, Leiduin/De Zilk vs. De Bilt) show a strong correlation (*Figure 2*). Assuming that this correlation is caused by a similar phenomenon, *i.e.* the long-range transport of γ–HCH, we are able to combine the time series of the De Bilt (1980-1999) with the time series of De Zilk (1999-2001). By doing so we obtain a long-term record of γ–HCH in precipitation in the Netherlands covering1980 to 2001. Moreover, continuation of the measurements in De Zilk will allow us to further extend this record of long-term measurements in the future. The frequency distribution of all measurements of the combined time series for De Bilt/De Zilk shows (*Figure 3*) most of the data to be approaching the limit of detection of the analytical method: 50 % of the data showing a concentration of 0.02 µg/l or less and 90 % a concentration of 0.1 µg/l or less. The arithmetic average concentration from this combined De Bilt/De Zilk data set, using a maximum likelihood estimator for a censored data set (Gilbert, 1987), is 0.04 µg/l.

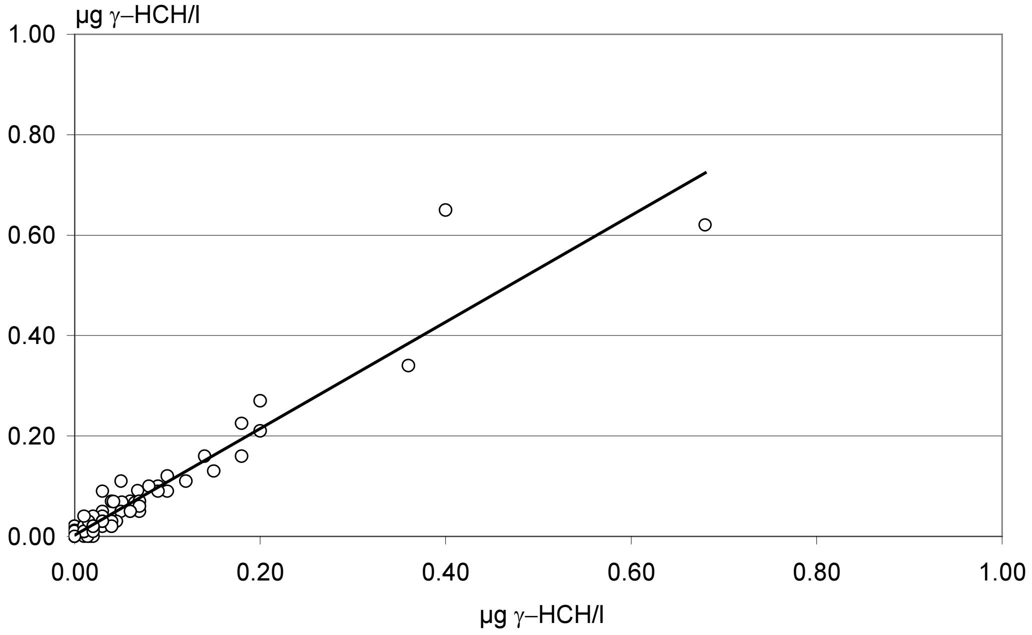

*Figure 2 Correlation between measurement results of γ–HCH in precipitation for Leiduin/De Zilk (horizontal) and De Bilt (vertical), 1988-1998. Regression equation y=1.06x + 0.003 with an explained variance of $R^2 = 0.92$*

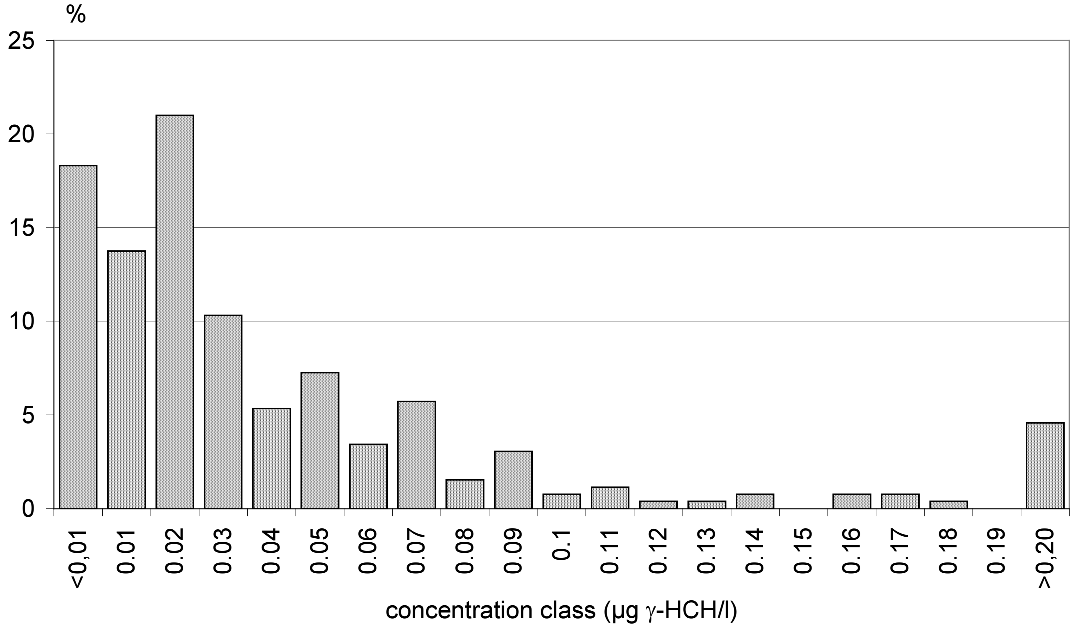

*Figure 3 Frequency distribution of measurement results of γ–HCH in precipitation for the combined time series of De Bilt/De Zilk. Total number of samples: 262.*

# 4. Discussion

## 4.1 Phenomenology

The strong correlation among the results of the various stations (compare *Figure 2*) supports the hypothesis that γ–HCH in Netherlands' precipitation is mainly the result of long-distance transport, as there are no local sources of lindane, and thus of γ–HCH, in the vicinity of the measurement sites. Peak values occur systematically every year somewhere in the time frame of April/June (*Figure 4*). This coincides with the use of lindane during this period of the year (Baart *et al*., 1995). For the rest of the year, concentrations were found to be around or below the detection limit of 0.01 µg/l. The same seasonal behaviour was observed at the EMEP monitoring sites in the nineties. Results of recent γ–HCH measurements in the EMEP network are summarised in *Table 3*. Average measurement results for the Netherlands from the mid-1990s are in the same range as the results of other stations in north-western Europe. Levels in remote areas (Ireland, Sweden, Iceland) come to a factor of ten (or more) lower. Peak values in the Netherlands, Germany and Belgium are relatively high and always occur in spring and/or early summer. Measurements results from Belgium and the Netherlands stations show the same seasonal behaviour of γ–HCH in precipitation (*Figure 5*). In general, the Belgian sites show the highest and the Netherlands' the lowest concentration and deposition values. The higher values in Belgium may most likely be attributed to the closer proximity of French source areas, where, until recently, lindane was being used extensively (A. van der Linden, personal communication).

*Table 3 Summarised results of selected γ–HCH measurements in precipitation in Europe, 1994-1997*

| Station Name | Year(s) | Median | Min | Max |
|---|---|---|---|---|
| | | µg/l | | |
| Knokke, Belgium | 1997 | 0.010 | 0.002 | 0.71 |
| Lista, Norway | 1996 | 0.008 | 0.002 | 0.025 |
| Rörvik, Sweden | 1996 | 0.010 | <0.001 | 0.027 |
| Stórhöfdi, Iceland | 1995-96 | <0.001 | <0.001 | 0.007 |
| Turlough Hill, Ireland | 1994-96 | 0.002 | <0.001 | 0.008 |
| Westerland, Germany | 1995-96 | 0.006 | <0.001 | 0.18 |
| Zingst, Germany | 1995-97 | 0.015 | <0.001 | 0.22 |
| De Bilt/De Zilk, Netherlands | 1994-97 | 0.015 | <0.010 | 0.23 |

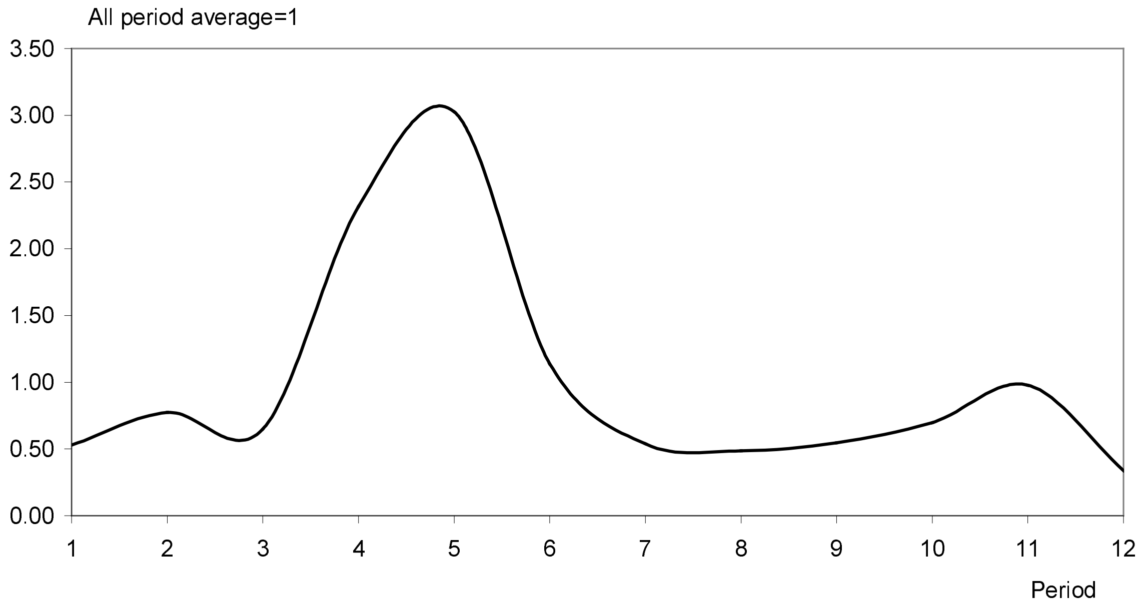

*Figure 4 Characteristic seasonal behaviour of γ–HCH concentrations in precipitation in the Netherlands. Overall period average is taken as 1. Periods '4' through '6' correspond roughly to the calendar period of April- June. Data for De Bilt/De Zilk, 1991-2000.*

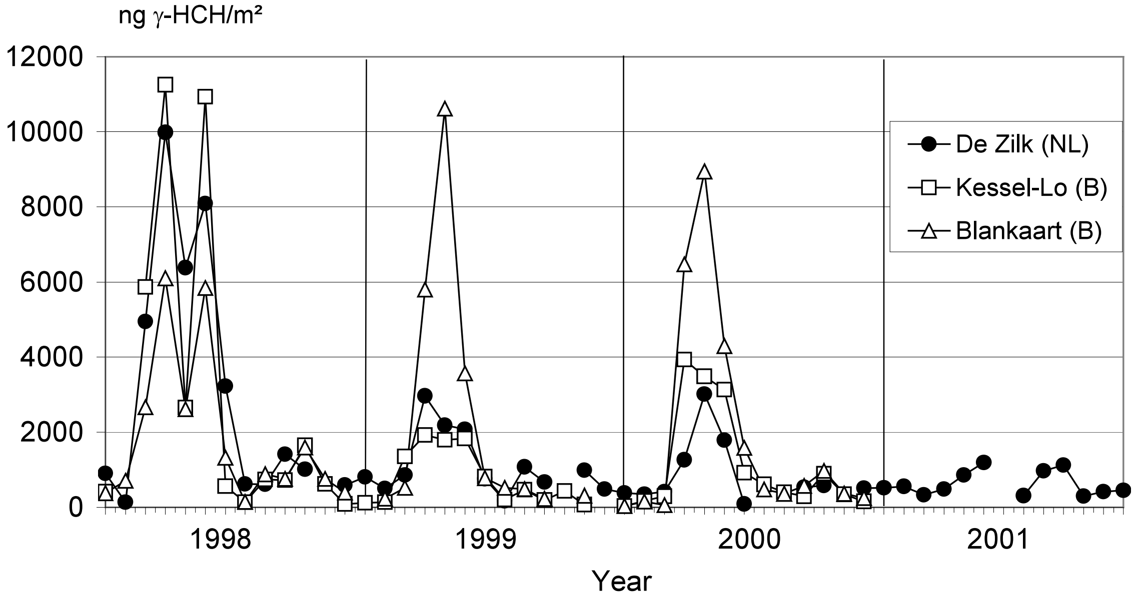

*Figure 5 Wet deposition of γ–HCH in precipitation on one of the Netherlands and two Belgian measurement sites (VMM, 1999, 2000, 2001), 1998-2001. Deposition data for 4-week periods.*

Further support for the role of long-range transport comes from the extensive research project on pesticides carried out in the Netherlands in 1999 and 2000 (Duyzer and Vonk, 2001). As part of this research, a network of 18 measurements sites was operated with the aim of describing the geographical distribution of a large number of pesticides in air and precipitation in the Netherlands. The concentrations of γ–HCH in precipitation at all the measurement sites show the same seasonal behaviour. Generally, concentrations throughout the year are low, with 75% of the samples showing a concentration below the limit of detection of the analytical method used (0.01 μg/l). Higher concentrations occur during a short period of time in spring. It was observed that these enhanced concentrations occur simultaneously throughout the network. Although there might be some influence of locally used lindane, this supports the hypothesis that a substantial part of γ–HCH concentrations in precipitation (and air) in the Netherlands can be attributed to long-range transport. Baart *et al.* (1995) carried out model calculations for the deposition of POP to the North Sea and the Netherlands. Major contributions to the γ–HCH deposition in the Netherlands come from France, Belgium and the Netherlands, with a contribution of 55, 20 and 15%, respectively. The emissions used in Baart *et al.* (1995) are a factor of 2-3 higher for the Netherlands and a factor of 2-3 lower for Belgium than those estimated by Breivik *et al.* (1999) [*Table 4*]. This means that the contribution of the Netherlands by Baart *et al.* (1995*)*, may probably be overestimated. Nevertheless, if only the emissions of the Netherlands were used in the calculations by Baart *et al.* (1995) this would lead to an underestimation of the observed concentration in the Netherlands by a factor of three. Using the more recent emission estimate by Breivik *et al.* (1999), the underestimation would even be a factor of seven. Although the uncertainty in the modelling of transport and deposition of lindane is quite large (about a factor of 2, see section 4.3), this clearly illustrates that observed levels of γ–HCH in the Netherlands can, to a large extent, be attributed to emissions by other countries *i.e.* France and Belgium.

*Tabel 4 Emissions of γ–HCH for the Netherlands, Belgium, France and Germany for 1980, 1985, 1990, 1996 in kg year$^{-1}$ taken from Breivik et al. (1999).*

| Country | 1980 | 1985 | 1990 | 1996 |
|---|---|---|---|---|
| Belgium | 19,075 | 23,049 | 15,411 | 15,890 |
| France | 322,228 | 294,072 | 270,456 | 560,000 |
| Germany | 111,447 | 49,608 | 27,662 | 0 |
| Netherlands | 32,686 | 36,589 | 3,814 | 6,307 |

## 4.2 A trend

Long-term measurement results may be suitable to revealing a long-term trend, not only in concentrations in precipitation but in emissions as well. The combined time series for De Bilt/De Zilk covering the 1980-2001 period has been used for this purpose. The median concentration value which is a robust measure for the central tendecy in a dataset, has been taken for each year (*Figure 6*) This time series shows a downward trend which is confirmed by the non-parametric Mann-Kendall test. The use of the complete dataset in stead of the median would problaby lead to erroreneous results. The use of the *seasonal* Mann-Kendall test seems approriate here. However, peak values do not always occur in the same sampling period. The result of the test then tends to be biased. Because of the shift of the peak values one could conclude wrongly that there is no trend. Data for the last three years -which are historically low- suggest a substantial and systematic decrease in lindane emissions in this period. Support for this hypothesis can be found in the measurements of γ–HCH in air in De Zilk in 1996 and 1999. According to Duyzer and Vonk (2001) concentrations of γ–HCH in precipitation have declined by a factor of three. Further measurements are required to confirm this hypothesis.

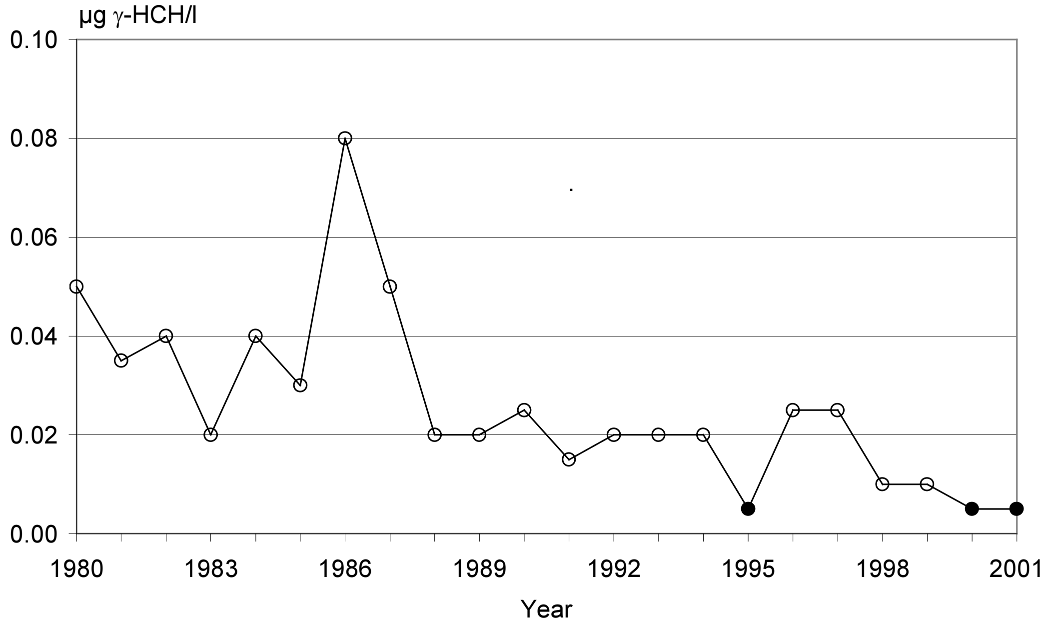

*Figure 6 Annual median concentrations of γ–HCH in precipitation in the Netherlands, 1980-2001. For the sake of convenience, median values which fall below the limit of the analytical method used (0.01 µg/l) has been arbitrarily goven the value of 0.005 µg/l. These data points are marked black (•).*

Recent emission inventories suggest that emission in Europe as a whole has declined from 2332 to 790 tons during the 1980 to 1996 period, i.e. a decrease of 75% (Holoubek *et al.*, 2000; see also *Table 4* for emissions of the Netherlands and surrounding countries). Nowadays, use of lindane is severely restricted or banned in most European countries. Until recently, major use of lindane was made in France and, to a lesser extent, Spain (Breivik *et al.*, 1999), with France accounting for 70% of the European emissions. The emissions of lindane in the Netherlands have strongly declined. The emissions at the end of the nineties were about a tenth of the emissions in the early eighties. The use of lindane has been prohibited in the Netherlands since 2000.

## 4.3 Comparison with results from models

A number of efforts to model the transport and deposition of lindane on a regional scale have been carried out (for a historical overview, see Van Jaarsveld and Van Pul, 1999). The models represent the spatial distribution of lindane over Europe reasonably well. The levels, however, show a sharp variation from the emission database used. When using two different emission estimates, measurements of lindane were both over- and underestimated by the

TREND model by a factor of 2-3, i.e. the two model calculations differed by near to a factor of 7 (Van Jaarsveld and Van Pul, 1999).

Recently, Shatalov *et al.* (2000) reported the modelling of lindane for the 1970-1997 period by the EMEP/MSC-E using the historical database of European lindane emissions by Breivik *et al.* (1999). Measurements were concluded to be, on average, twice as high as the calculations (for roughly the same stations as in *Table 3*). For individual stations, modelling results differ from 0.5 to 10 times the measured values. The differences can be largely attributed to the uncertainties in the emission estimates. Besides, a systematic deviation through modelling may be present, as a result of uncertainties in the removal processes parameterisation (chemical degradation and deposition) and the physicochemical properties used in the parameterisation. For pesticides, this was estimated at a factor of 3 (Van Pul *et al.*, 1999). For lindane, a fairly well-known component, this may be somewhat better, *e.g.* factor of 2. The emission estimates are too coarse in time to be able to evaluate explicitly the year-to-year variation in the lindane concentrations in precipitation.

# 5. Conclusions

The long-term measurement of γ-HCH in precipitation in the Netherlands covers the period from 1980 to the present. Concentrations of γ-HCH in precipitation show a systematic seasonal behaviour, with generally low concentrations throughout the year. During a short period in April/June concentrations rise. There is evidence that γ-HCH concentrations in the precipitation in the Netherlands have significantly decreased. Moreover, measurement results from the last three years (1999-2001) show a clear downward trend as well and suggest a decline in lindane emissions.

Measurement results from neighbouring countries shows a similar pattern: enhanced concentrations in precipitation during the same period. This supports the hypothesis γ-HCH in precipitation is a large-scale occurrence. Major sources of γ-HCH in precipitation in the Netherlands (and the neighbouring countries) are most probably source areas in France, where, until recently, lindane was used extensively.

The large uncertainties in emission estimates, in atmospheric transport and in deposition modelling complicate the analysis of the state of the environmental levels and the change in time. Measurements are in this case probably a better tool to evaluate environmental levels of γ–HCH.

# Acknowledgements

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# Appendix 1 Mailing list

1. Mw Mr T Zwartpoorte, Directeur Klimaatverandering en Industrie
2. Drs WF Iestra, Directie Klimaatverandering en Industrie
3. Ir JA Herremans, Directie Lokale Milieukwaliteit en Verkeer
4. Mr AB Holtkamp, Directie Stoffen, Afvalstoffen, Straling
5. Dr H Meyer, Directie Bodem, Water, Landelijk Gebied
6. Drs CJ Sliggers, Directie Klimaatverandering en Industrie
7. Dr WAH Asman, Danish Institute of Agricultural Sciences
8. Dr E Brorström-Lundén, Swedish Environmental Research Institute
9. Dr P Coleman, AEA Technology, National Environmental Technology Centre
10. Dr S Dutchak, EMEP/MSC-E
11. Dr JH Duyzer, TNO
12. Dr I Holoubek, RECETOX
13. Dr K Jones, Lancaster University
14. Dr G Lammel, MPI für Meteorologie
15. Dr D Mackay, Trent University
16. Dr A Malaninchev, EMEP/MSC-E
17. Dr S Mannø, Norwegian Institute for Air Research, NILU
18. Dr J Munthe, IVL
19. Dr J Pacyna, Norwegian Institute for Air Research, NILU
20. Dr A Sweetman, Lancaster University
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24. Ir JP Beck
25. Ir HSMA Diederen
26. Drs HC Eerens
27. Prof Ir ND van Egmond
28. Ir FJM Folkert
29. Dr ing JA van Jaarsveld
30. Drs JA Janus
31. Dr FAAM de Leeuw
32. Ir AMA van der Linden
33. Dr ir D van de Meent
34. Dr W Slooff
35. Ir AP Stolk
36. Drs K van Velze
37. Ir K Wieringa
38. Drs E Buijsman
39. Dr WAJ van Pul
40. SBC/Communicatie
41. Bureau Rapportenregistratie
42. Bibliotheek RIVM

43-48. Bureau Rapportenbeheer

49-60. Reserve exemplaren

# Appendix 2 Measurement results

The following tables present the measurements results of γ-HCH in precipitation (‘c’; in µg γ-HCH /l) as well as the amount of precipitation (‘mm’, in mm). The length of the sampling period was one month (until 1991) or four weeks (from 1991 onwards).

Na: not analysed

Ns: no sample

*Tabel A2.1 Measurement data for De Bilt, 1980-1985.*

| Year | Period | mm | c | Year | Period | mm | c |
|---|---|---|---|---|---|---|---|
| **1980** | 80/1 | 19 | 0.04 | **1983** | 83/1 | 87 | 0.01 |
| | 80/2 | 56 | 0.04 | | 83/2 | 60 | 0.02 |
| | 80/3 | 114 | 0.08 | | 83/3 | 89 | Ns |
| | 80/4 | 60 | 0.05 | | 83/4 | 106 | 0.14 |
| | 80/5 | 18 | 0.23 | | 83/5 | 118 | 0.18 |
| | 80/6 | 120 | 0.08 | | 83/6 | 35 | 0.17 |
| | 80/7 | 37 | 0.02 | | 83/7 | 25 | 0.06 |
| | 80/8 | 73 | 0.05 | | 83/8 | 33 | 0.04 |
| | 80/9 | 59 | 0.05 | | 83/9 | 121 | 0.02 |
| | 80/10 | 86 | 0.05 | | 83/10 | 40 | 0.01 |
| | 80/11 | 89 | 0.07 | | 83/11 | 88 | 0.01 |
| | 80/12 | 125 | 0.03 | | 83/12 | 105 | 0.01 |
| **1981** | 81/1 | 114 | 0.03 | **1984** | 84/1 | 147 | 0.02 |
| | 81/2 | 39 | 0.04 | | 84/2 | 73 | 0.01 |
| | 81/3 | 134 | 0.03 | | 84/3 | 65 | 0.02 |
| | 81/4 | 16 | 0.06 | | 84/4 | 14 | 0.03 |
| | 81/5 | 96 | 0.22 | | 84/5 | 90 | 0.07 |
| | 81/6 | 60 | 0.02 | | 84/6 | 62 | 0.06 |
| | 81/7 | 105 | <0.01 | | 84/7 | 74 | 0.05 |
| | 81/8 | 30 | 0.01 | | 84/8 | 9 | 0.05 |
| | 81/9 | 52 | 0.05 | | 84/9 | 148 | 0.02 |
| | 81/10 | 189 | 0.03 | | 84/10 | 116 | 0.06 |
| | 81/11 | 59 | 0.09 | | 84/11 | 65 | 0.08 |
| | 81/12 | 99 | 0.02 | | 84/12 | 29 | 0.03 |
| **1982** | 82/1 | 45 | 0.04 | **1985** | 85/1 | 35 | 0.05 |
| | 82/2 | 16 | 0.09 | | 85/2 | 6 | Na |
| | 82/3 | 62 | 0.03 | | 85/3 | 58 | 0.02 |
| | 82/4 | 32 | 0.11 | | 85/4 | 81 | 0.09 |
| | 82/5 | 26 | 0.14 | | 85/5 | 40 | 0.05 |
| | 82/6 | 91 | 0.04 | | 85/6 | 103 | 0.04 |
| | 82/7 | 27 | 0.02 | | 85/7 | 83 | 0.02 |
| | 82/8 | 51 | 0.02 | | 85/8 | 78 | 0.02 |
| | 82/9 | 25 | 0.05 | | 85/9 | 51 | 0.02 |
| | 82/10 | 109 | 0.04 | | 85/10 | 35 | 0.01 |
| | 82/11 | 70 | 0.03 | | 85/11 | 16 | 0.03 |
| | 82/12 | 80 | 0.02 | | 85/12 | 86 | 0.05 |

*>>>continued on next page*

*Tabel A2.1 (continued) Measurement data for De Bilt, 1986-1991.*

| Year | Period | mm | c | Year | Period | mm | c |
|---|---|---|---|---|---|---|---|
| **1986** | 86/1 | 112 | 0.01 | **1989** | 89/1 | 31 | 0.02 |
| | 86/2 | 1 | Na | | 89/2 | 48 | 0.02 |
| | 86/3 | 82 | Ns | | 89/3 | 119 | 0.02 |
| | 86/4 | 32 | 0.49 | | 89/4 | 76 | 0.16 |
| | 86/5 | 52 | 2.00 | | 89/5 | 6 | Na |
| | 86/6 | 45 | 0.24 | | 89/6 | 67 | 0.04 |
| | 86/7 | 45 | 0.07 | | 89/7 | 32 | 0.03 |
| | 86/8 | 84 | 0.07 | | 89/8 | 36 | <0.01 |
| | 86/9 | 26 | 0.05 | | 89/9 | 93 | 0.03 |
| | 86/10 | 137 | 0.01 | | 89/10 | 84 | <0.01 |
| | 86/11 | 89 | 0.27 | | 89/11 | 36 | <0.01 |
| | 86/12 | 118 | 0.09 | | 89/12 | 90 | 0.03 |
| | | | | | | | |
| **1987** | 87/1 | 38 | Ns | **1990** | 90/1 | 58 | 0.02 |
| | 87/2 | 7 | 0.02 | | 90/2 | 102 | 0.02 |
| | 87/3 | 61 | <0.01 | | 90/3 | 58 | 0.05 |
| | 87/4 | 61 | 0.17 | | 90/4 | 52 | 0.21 |
| | 87/5 | 88 | 0.06 | | 90/5 | 32 | 0.10 |
| | 87/6 | 78 | 0.07 | | 90/6 | 70 | 0.05 |
| | 87/7 | 17 | 0.05 | | 90/7 | 28 | 0.02 |
| | 87/8 | 84 | 0.02 | | 90/8 | 54 | <0.01 |
| | 87/9 | 62 | 0.03 | | 90/9 | 96 | 0.02 |
| | 87/10 | 104 | 0.06 | | 90/10 | 70 | 0.03 |
| | 87/11 | 10 | 0.07 | | 90/11 | 102 | 0.03 |
| | 87/12 | 61 | 0.02 | | 90/12 | 90 | 0.01 |
| | | | | | | | |
| **1988** | 88/1 | 126 | 0.01 | **1991** | 91/1 | 26 | 0.02 |
| | 88/2 | 92 | <0.01 | | 91/2 | 23 | 0.01 |
| | 88/3 | 122 | 0.02 | | 91/3 | 22 | 0.07 |
| | 88/4 | 19 | 0.62 | | 91/4 | 32 | 0.07 |
| | 88/5 | 43 | 0.65 | | 91/5 | 22 | 0.03 |
| | 88/6 | 13 | 0.12 | | 91/6 | 81 | 0.09 |
| | 88/7 | 154 | 0.07 | | 91/7 | 131 | 0.07 |
| | 88/8 | 63 | 0.02 | | 91/8 | 18 | 0.02 |
| | 88/9 | 89 | 0.02 | | 91/9 | 2 | 0.02 |
| | 88/10 | 60 | 0.02 | | 91/10 | 91 | <0.01 |
| | 88/11 | 50 | 0.07 | | 91/11 | 85 | 0.02 |
| | 88/12 | 80 | 0.01 | | 91/12 | 109 | 0.02 |
| | | | | | 91/13 | 55 | 0.02 |

*>>>continued on next page*

*Tabel A2.1 (continued) Measurement data for De Bilt, 1992-1997.*

| Year | Period | mm | c | Year | Period | mm | c |
|---|---|---|---|---|---|---|---|
| **1992** | 92/1 | 37 | 0.02 | **1995** | 95/1 | 127 | <0.01 |
| | 92/2 | 39 | 0.02 | | 95/2 | 171 | <0.01 |
| | 92/3 | 76 | 0.07 | | 95/3 | 107 | 0.01 |
| | 92/4 | 42 | 0.07 | | 95/4 | 59 | 0.04 |
| | 92/5 | 57 | 0.34 | | 95/5 | 21 | 0.03 |
| | 92/6 | 58 | 0.06 | | 95/6 | 96 | 0.11 |
| | 92/7 | 59 | 0.03 | | 95/7 | 46 | 0.02 |
| | 92/8 | 51 | 0.02 | | 95/8 | 55 | <0.01 |
| | 92/9 | 170 | 0.01 | | 95/9 | 24 | <0.01 |
| | 92/10 | 12 | 0.02 | | 95/10 | 116 | <0.01 |
| | 92/11 | 134 | 0.02 | | 95/11 | 30 | 0.03 |
| | 92/12 | 127 | 0.02 | | 95/12 | 60 | 0.01 |
| | 92/13 | 61 | 0.01 | | 95/13 | 30 | 0.02 |
| **1993** | 93/1 | 94 | 0.02 | **1996** | 96/1 | 8 | 0.02 |
| | 93/2 | 7 | 0.01 | | 96/2 | 78 | 0.01 |
| | 93/3 | 36 | 0.05 | | 96/3 | 21 | 0.01 |
| | 93/4 | 49 | 0.09 | | 96/4 | 22 | 0.05 |
| | 93/5 | 19 | 0.16 | | 96/5 | 9 | 0.09 |
| | 93/6 | 63 | 0.05 | | 96/6 | 71 | 0.1 |
| | 93/7 | 80 | 0.02 | | 96/7 | 68 | 0.03 |
| | 93/8 | 133 | <0.01 | | 96/8 | 46 | 0.02 |
| | 93/9 | 37 | <0.01 | | 96/9 | 57 | 0.02 |
| | 93/10 | 129 | 0.01 | | 96/10 | 84 | 0.02 |
| | 93/11 | 43 | 0.02 | | 96/11 | 27 | 0.03 |
| | 93/12 | | Ns | | 96/12 | 199 | 0.03 |
| | 93/13 | | Ns | | 96/13 | | Ns |
| **1994** | 94/1 | 85 | <0.01 | **1997** | 97/1 | | |
| | 94/2 | 24 | <0.01 | | 97/2 | 90 | 0.02 |
| | 94/3 | 83 | 0.02 | | 97/3 | 32 | 0.02 |
| | 94/4 | 96 | 0.04 | | 97/4 | 26 | 0.11 |
| | 94/5 | 42 | 0.225 | | 97/5 | 104 | 0.27 |
| | 94/6 | 109 | 0.09 | | 97/6 | 93 | 0.13 |
| | 94/7 | 10 | 0.02 | | 97/7 | 98 | 0.03 |
| | 94/8 | 68 | 0.03 | | 97/8 | 60 | <0.01 |
| | 94/9 | 92 | 0.02 | | 97/9 | 116 | 0.01 |
| | 94/10 | 166 | 0.01 | | 97/10 | 36 | 0.02 |
| | 94/11 | 102 | 0.03 | | 97/11 | 49 | 0.04 |
| | 94/12 | 39 | 0.04 | | 97/12 | 26 | 0.03 |
| | 94/13 | 111 | 0.02 | | 97/13 | 80 | 0.02 |

*Tabel A2.2 Measurement data for Witteveen, 1985-1987.*

| Year | Period | mm | c |
|---|---|---|---|
| **1985** | 85/1 | 65 | <0.01 |
| | 85/2 | 7 | Na |
| | 85/3 | 75 | <0.01 |
| | 85/4 | 75 | 0.08 |
| | 85/5 | 19 | 0.04 |
| | 85/6 | 105 | 0.03 |
| | 85/7 | 97 | <0.01 |
| | 85/8 | 102 | 0.01 |
| | 85/9 | 61 | 0.01 |
| | 85/10 | 28 | 0.02 |
| | 85/11 | 82 | 0.04 |
| | 85/12 | 89 | 0.03 |
| | | | |
| **1986** | 86/1 | 116 | 0.01 |
| | 86/2 | 4 | Na |
| | 86/3 | 69 | Ns |
| | 86/4 | 39 | 0.03 |
| | 86/5 | 29 | 1.00 |
| | 86/6 | 70 | 0.33 |
| | 86/7 | 44 | 0.06 |
| | 86/8 | 44 | 0.07 |
| | 86/9 | 42 | 0.05 |
| | 86/10 | 89 | 0.09 |
| | 86/11 | 65 | 0.17 |
| | 86/12 | 116 | 0.07§ |
| | | | |
| **1987** | 87/1 | 68 | Ns |
| | 87/2 | 76 | 0.01 |
| | 87/3 | 51 | 0.02 |
| | 87/4 | 21 | 0.10 |
| | 87/5 | 83 | 0.10 |
| | 87/6 | 101 | 0.05 |
| | 87/7 | 102 | 0.04 |
| | 87/8 | 65 | 0.01 |
| | 87/9 | 70 | 0.01 |
| | 87/10 | 80 | 0.04 |
| | 87/11 | 90 | 0.02 |
| | 87/12 | 84 | 0.02 |

§ Incomplete sample

*Tabel A2.3 Measurement data for Vlissingen, 1985-1987.*

| Year | Period | mm | c |
|---|---|---|---|
| **1985** | 85/1 | 30 | 0.03 |
| | 85/2 | 1 | Na |
| | 85/3 | 77 | <0.01 |
| | 85/4 | 61 | 0.16 |
| | 85/5 | 60 | 0.06 |
| | 85/6 | 81 | 0.04 |
| | 85/7 | 59 | 0.02 |
| | 85/8 | 60 | 0.01 |
| | 85/9 | 22 | 0.01 |
| | 85/10 | 45 | 0.02 |
| | 85/11 | 76 | 0.05 |
| | 85/12 | 69 | 0.01 |
| | | | |
| **1986** | 86/1 | 104 | 0.01 |
| | 86/2 | 1 | Na |
| | 86/3 | 84 | Na |
| | 86/4 | 44 | 0.59 |
| | 86/5 | 42 | 2.80 |
| | 86/6 | 62 | 0.27 |
| | 86/7 | 24 | 0.18 |
| | 86/8 | 121 | 0.10 |
| | 86/9 | 46 | 0.05 |
| | 86/10 | 91 | 0.14 |
| | 86/11 | 76 | 0.31 |
| | 86/12 | 90 | 0.08 |
| | | | |
| **1987** | 87/1 | 31 | |
| | 87/2 | 31 | 0.02 |
| | 87/3 | 74 | 0.05 |
| | 87/4 | 2 | 0.44 |
| | 87/5 | 85 | 0.19 |
| | 87/6 | 123 | 0.05 |
| | 87/7 | 108 | 0.02 |
| | 87/8 | 77 | 0.02 |
| | 87/9 | 55 | 0.02 |
| | 87/10 | 85 | 0.06 |
| | 87/11 | 96 | 0.06 |
| | 87/12 | 29 | 0.06 |

*Tabel A2.4 Measurement data for Leiduin, 1988-1993.*

| Year | Period | mm | c | Year | Period | mm | c |
|---|---|---|---|---|---|---|---|
| **1988** | 88/1 | 133 | 0.01 | **1991** | 91/1 | 28 | 0.01 |
| | 88/2 | 60 | <0.01 | | 91/2 | 17 | <0.01 |
| | 88/3 | 107 | 0.20 | | 91/3 | 32 | 0.05 |
| | 88/4 | 19 | 0.68 | | 91/4 | 36 | 0.04 |
| | 88/5 | 24 | 0.40 | | 91/5 | 31 | 0.02 |
| | 88/6 | 6 | 0.10 | | 91/6 | 46 | 0.07 |
| | 88/7 | 99 | 0.06 | | 91/7 | 125 | 0.07 |
| | 88/8 | 117 | 0.02 | | 91/8 | 18 | 0.02 |
| | 88/9 | 105 | 0.01 | | 91/9 | 17 | 0.01 |
| | 88/10 | 63 | 0.02 | | 91/10 | 105 | <0.01 |
| | 88/11 | 64 | 0.04 | | 91/11 | 68 | 0.02 |
| | 88/12 | 64 | 0.01 | | 91/12 | 85 | 0.02 |
| | | | | | 91/13 | 43 | 0.01 |
| **1989** | 89/1 | 20 | 0.01 | **1992** | 92/1 | 49 | 0.02 |
| | 89/2 | 50 | 0.03 | | 92/2 | 26 | 0.02 |
| | 89/3 | 75 | <0.01 | | 92/3 | 57 | 0.07 |
| | 89/4 | 76 | 0.14 | | 92/4 | 42 | 0.07 |
| | 89/5 | 8 | Na | | 92/5 | 39 | 0.36 |
| | 89/6 | 89 | 0.03 | | 92/6 | 44 | 0.07 |
| | 89/7 | 44 | 0.04 | | 92/7 | 68 | 0.02 |
| | 89/8 | 50 | <0.01 | | 92/8 | 41 | 0.02 |
| | 89/9 | 59 | 0.04 | | 92/9 | 147 | 0.01 |
| | 89/10 | 83 | <0.01 | | 92/10 | 104 | 0.02 |
| | 89/11 | 48 | <0.01 | | 92/11 | 150 | 0.02 |
| | 89/12 | 72 | 0.03 | | 92/12 | 111 | 0.02 |
| | | | | | 92/13 | 60 | 0.01 |
| **1990** | 90/1 | 41 | 0.02 | **1993** | 93/1 | 62 | 0.01 |
| | 90/2 | 83 | <0.01 | | 93/2 | 20 | 0.01 |
| | 90/3 | 44 | 0.03 | | 93/3 | 14 | 0.06 |
| | 90/4 | 45 | 0.20 | | 93/4 | 33 | 0.10 |
| | 90/5 | 20 | 0.09 | | 93/5 | 49 | 0.18 |
| | 90/6 | 80 | 0.07 | | 93/6 | . | 0.05 |
| | 90/7 | 6 | Na | | 93/7 | . | 0.02 |
| | 90/8 | 71 | 0.01 | | 93/8 | | |
| | 90/9 | 95 | 0.01 | | 93/9 | | |
| | 90/10 | 57 | 0.03 | | 93/10 | | |
| | 90/11 | 128 | 0.03 | | 93/11 | | |
| | 90/12 | 56 | 0.01 | | 93/12 | | |
| | | | | | 93/13 | | |

*Tabel A2.5 Measurement data for Rotterdam, 1988-1991.*

| Year | Period | mm | c | Year | Period | mm | c |
|---|---|---|---|---|---|---|---|
| **1988** | 88/1 | 127 | 0.01 | **1990** | 90/1 | 55 | 0.02 |
| | 88/2 | 71 | <0.01 | | 90/2 | 96 | 0.02 |
| | 88/3 | 121 | 0.02 | | 90/3 | 50 | 0.04 |
| | 88/4 | 16 | 0.68 | | 90/4 | 51 | 0.24 |
| | 88/5 | 34 | 0.55 | | 90/5 | 28 | 0.09 |
| | 88/6 | 19 | 0.18 | | 90/6 | 61 | 0.07 |
| | 88/7 | 154 | 0.07 | | 90/7 | 28 | <0.01 |
| | 88/8 | 63 | 0.02 | | 90/8 | 51 | 0.02 |
| | 88/9 | 89 | 0.02 | | 90/9 | 108 | 0.02 |
| | 88/10 | 81 | 0.03 | | 90/10 | 52 | 0.03 |
| | 88/11 | 55 | 0.08 | | 90/11 | 97 | 0.03 |
| | 88/12 | 65 | 0.02 | | 90/12 | 60 | 0.02 |
| | | | | | | | |
| **1989** | 89/1 | 23 | 0.02 | **1991** | 91/1 | 39 | 0.01 |
| | 89/2 | 55 | 0.02 | | 91/2 | 25 | 0.01 |
| | 89/3 | 94 | <0.01 | | 91/3 | 32 | 0.07 |
| | 89/4 | 62 | 0.19 | | 91/4 | 24 | 0.10 |
| | 89/5 | 9 | Na | | 91/5 | 42 | 0.05 |
| | 89/6 | 81 | 0.06 | | 91/6 | 58 | 0.10 |
| | 89/7 | 24 | 0.04 | | 91/7 | 169 | 0.06 |
| | 89/8 | 47 | 0.01 | | 91/8 | 16 | 0.02 |
| | 89/9 | 51 | 0.03 | | 91/9 | 10 | 0.02 |
| | 89/10 | 90 | <0.01 | | 91/10 | 92 | 0.01 |
| | 89/11 | 44 | <0.01 | | 91/11 | 84 | 0.02 |
| | 89/12 | 88 | 0.03 | | 91/12 | 87 | 0.02 |
| | | | | | 91/13 | 36 | 0.02 |

*Tabel A2.6 Measurement data for De Zilk, 1994-1999.*

| Year | Period | mm | c | Year | Period | mm | c |
|---|---|---|---|---|---|---|---|
| **1994** | | | | **1997** | 97/1 | | |
| | | | | | 97/2 | 33 | 0.02 |
| | | | | | 97/3 | 41 | 0.04 |
| | | | | | 97/4 | 23 | 0.12 |
| | 94/5 | 36 | 0.18 | | 97/5 | 59 | 0.20 |
| | 94/6 | 37 | 0.09 | | 97/6 | 46 | 0.15 |
| | 94/7 | 21 | 0.04 | | 97/7 | 74 | 0.03 |
| | 94/8 | 60 | 0.02 | | 97/8 | 66 | <0.01 |
| | 94/9 | 80 | 0.02 | | 97/9 | 87 | 0.02 |
| | 94/10 | 148 | <0.01 | | 97/10 | 36 | 0.02 |
| | 94/11 | 126 | 0.02 | | 97/11 | 158 | 0.01 |
| | 94/12 | 33 | 0.03 | | 97/12 | 44 | 0.03 |
| | 94/13 | 105 | 0.01 | | 97/13 | 94 | 0.02 |
| | | | | | | | |
| **1995** | 95/1 | 121 | <0.01 | **1998** | 98/1 | 181 | 0.01 |
| | 95/2 | 125 | <0.01 | | 98/2 | 13 | 0.01 |
| | 95/3 | 110 | <0.01 | | 98/3 | 165 | 0.03 |
| | 95/4 | 51 | 0.02 | | 98/4 | 111 | 0.09 |
| | 95/5 | 20 | 0.05 | | 98/5 | 91 | 0.07 |
| | 95/6 | 93 | 0.05 | | 98/6 | 101 | 0.08 |
| | 95/7 | 88 | 0.01 | | 98/7 | 161 | 0.02 |
| | 95/8 | 45 | <0.01 | | 98/8 | 123 | <0.01 |
| | 95/9 | 56 | <0.01 | | 98/9 | 122 | <0.01 |
| | 95/10 | 109 | <0.01 | | 98/10 | 282 | <0.01 |
| | 95/11 | 21 | 0.03 | | 98/11 | 202 | <0.01 |
| | 95/12 | 63 | <0.01 | | 98/12 | | Ns |
| | 95/13 | 34 | 0.01 | | 98/13 | 117 | 0.01 |
| | | | | | | | |
| **1996** | 96/1 | 14 | 0.01 | **1999** | 99/1 | 79 | 0.01 |
| | 96/2 | 46 | 0.01 | | 99/2 | 53 | 0.01 |
| | 96/3 | 24 | 0.01 | | 99/3 | 87 | 0.01 |
| | 96/4 | 25 | 0.06 | | 99/4 | 72 | 0.04 |
| | 96/5 | 11 | 0.03 | | 99/5 | 34 | 0.06 |
| | 96/6 | 48 | 0.08 | | 99/6 | 66 | 0.03 |
| | 96/7 | 62 | 0.02 | | 99/7 | 71 | 0.01 |
| | 96/8 | 37 | 0.02 | | 99/8 | 14 | 0.01 |
| | 96/9 | 155 | 0.02 | | 99/9 | 104 | 0.01 |
| | 96/10 | 65 | 0.02 | | 99/10 | 59 | 0.01 |
| | 96/11 | 118 | 0.04 | | 99/11 | 138 | <0.01 |
| | 96/12 | 131 | 0.03 | | 99/12 | 98 | 0.01 |
| | 96/13 | 68 | 0.02 | | 99/13 | 92 | 0.01 |

>>>*continued on next page*

*Tabel A2.6 (continued) Measurement data for De Zilk, 2000-2001.*

| Year | Period | mm | c |
|---|---|---|---|
| **2000** | 00/1 | 77 | <0.01 |
| | 00/2 | 70 | <0.01 |
| | 00/3 | 84 | <0.01 |
| | 00/4 | 21 | 0.06 |
| | 00/5 | 43 | 0.07 |
| | 00/6 | 89 | 0.02 |
| | 00/7 | 18 | <0.01 |
| | 00/8 | 49 | Ns |
| | 00/9 | 15 | 0.01 |
| | 00/10 | 107 | <0.01 |
| | 00/11 | 115 | <0.01 |
| | 00/12 | 200 | Ns |
| | 00/13 | 101 | <0.01 |
| | | | |
| **2001** | 00/1 | 104 | <0.01 |
| | 00/2 | 111 | <0.01 |
| | 00/3 | 67 | <0.01 |
| | 00/4 | 97 | <0.01 |
| | 00/5 | 86 | <0.01 |
| | 00/6 | 59 | 0.02 |
| | 00/7 | 37 | <0.01 |
| | 00/8 | 62 | <0.01 |
| | 00/9 | 194 | <0.01 |
| | 00/10 | 224 | <0.01 |
| | 00/11 | 59 | <0.01 |
| | 00/12 | 83 | <0.01 |
| | 00/13 | 89 | <0.01 |