MicroNAC

Advanced tools for assessment of the <u>Micro</u>bial <u>N</u>atural <u>A</u>ttenuation <u>C</u>apacity for <u>Micro</u>pollutants in the water cycle

Project duration: December 2013-December 2016

Project partners: Department Environmental Technology, Wageningen University (ETE) Laboratory of Microbiology, Wageningen University (MIB) Water Laboratorium Noord (WLN) Vitens

1. Executive Summary (max 1 page):

The increasing presence of organic micropollutants in different segments of the water cycle will or can threaten future water resources. These micropollutants are currently being detected at low concentrations in groundwater and surface water used for drinking water. While current monitoring (chemical analyses) gives an indication of the presence of these micropollutants, little is known about the natural attenuation of micropollutants under in situ conditions. There is a lack of information on biodegradation rates under environmental conditions and a necessity for more tools to assess natural attenuation capacity. This information is required to improve models used to assess and predict the long term risks of contamination of drinking water intakes.

This research project proposed here aims to improve the understanding of biodegradation of micropollutants by developing tools to determine biodegradation capacity of a number of key micropollutants, resulting in assessment and prediction guidelines for natural attenuation. The project will focus on a number of micropollutants that specifically threaten Dutch drinking water quality. For these compounds, biomolecular tools based on DNA analysis will be setup to assess the natural attenuation capacity in field samples. Additionally, ex situ degradation experiments using field samples as inoculum will be used to estimate degradation rates. Results will be integrated to form guidelines for the prediction of natural attenuation using molecular tools.

The project will be performed as a collaboration between private sector drinking water companies and Wageningen University. The drinking water companies Vitens, WMD and Waterbedrijf Groningen (the latter two represented by WLN) and will provide expertise on drinking water practices and the current risks to these intakes as well as monitoring results and ground- and surface water samples. At Wageningen University, work will be executed within the department of Environmental Technology and the Laboratory of Microbiology. Research will focus on developing and applying tools that can be used to assess the in situ natural attenuation capacity of specific micropollutants. The integration of more fundamental research at the university level with practical knowledge by drinking water companies will be used to develop and establish guidelines for assessing in situ natural attenuation capacity.

This project fits within the Topsector Water's focus on "Water for All" by developing sustainable, cost-effective practices to ensure the future and water quality of Dutch drinking water resources. By establishing tools to assess natural attenuation capacity and degradation rates, this information can be incorporated into risk assessment models used to predict the future security of drinking water resources. The economic and social benefits of clean source drinking water practices are numerous.

2. Background (max 3 pages)

Motivation and connection with Topsector Water, specifically with the "Innovatiecontract Watertechnologie"

Proper control of water quality from the source to the tap is essential to ensure the security of Dutch drinking water. Good management means that the supplied water complies with legal requirements and is perceived as a clear, tasteless, odorless, and healthy product by the consumer. This requires a good understanding of the extracted water, a properly controlled purification and distribution system, and adequate quality controls. Regular monitoring of extracted water, be it surface or groundwater, ensures the quality of source water. Additionally, monitoring of groundwater near extraction wells gives insight into the long-term quality of source water.

The presence of organic micropollutants in the environment threatens surface and groundwater intakes for drinking water. Increasingly, organic micropollutants have been measured during routine monitoring of source and groundwater, which is of concern for both current and future drinking water resources. While often encountered at low concentrations (<10 μ g/L), the environmental fate, persistence and eco-toxicological risk of compounds such as pesticides, pharmaceuticals, personal care products, hormones, and flame retardants is often unknown. In order to ensure the safety of future drinking water resources, this project investigates the natural attenuation capacity of microorganisms to degrade a number of the most problematic compounds. This research aims to identify degraders and establish monitoring techniques using molecular tools to assess the potential for biodegradation.

The results of this project are essential to securing drinking water resources, a high priority within Topsector Water's theme "Water for all". Here, focus is set on the "security of production of future drinking and industry water and advanced waste water treatment techniques", with innovative technologies aiming at safer, more sustainable, and more efficient processes. The development of tools to assess natural attenuation of organic micropollutants is essential to ensure the safety of drinking water resources. Reliance on clean drinking water resources ("schone bron") and natural attenuation processes is more sustainable than implementing predistribution purification techniques of drinking water with costly infrastructure. Additionally, while the focus of this research is on organic micropollutants in drinking water resources, the tools developed are applicable to other portions of the water cycle, such as waste water treatment.

Innovation/technology

Current monitoring of water resources gives an indication of the presence of organic micropollutants. New analytical techniques are available to determine the concentration of micropollutants and some transformation products. However, risk assessment using models is difficult due to the shortage of information on natural attenuation of these compounds. If some natural attenuation or biodegradation can be demonstrated, the risks associated with micropollutants can be affected. For many micropollutants, there lacks information on biodegradation rates under in situ environmental conditions or ways to assess biodegradation capacity. Laboratory based degradation experiments are often performed in a matter of weeks to months under optimal conditions. It is difficult to translate the results of these experiments to field conditions, where residence times between monitoring and intake wells may be much longer (10-15 years for groundwater reservoirs) and suboptimal degradation conditions are encountered.

This research project aims to improve the understanding of biodegradation of organic micropollutants by developing tools to determine biodegradation capacity for a number of micropollutants. Biomolecular tools using DNA analyses can effectively and reliably monitor the presence and abundance of microorganisms and their genes in the environment. Biomolecular assays have been extensively applied for the determination of biodegradation capacity at contaminated locations, for example, for reductive dechlorination of chlorinated ethenes. While some molecular tools are available for determining the presence of degraders or genes involved in degradation of micropollutants, the field is still in its infancy. Significant further research is required to identify degraders and develop and validate assays to monitor their abundance. The research proposed here will contribute fundamental scientific knowledge on biodegradation of organic micropollutants, which can be used in multiple areas within the water cycle.

This project will contribute to improved assessment tools and prediction guidelines for natural attenuation of micropollutants that specifically threaten Dutch drinking water resources. We aim to match the advances in analysis of the quantification of micropollutants with tools to assess the natural attenuation of these compounds. The combination of proper analytics with novel tools for monitoring biodegradation is necessary to ensure future drinking water resources.

Economic perspective/market potential

Contamination of drinking water sources in the future could cause significant societal costs. The results of this project will improve our capacity to predict the future quality of drinking water source water. This is essential to our ability to ensure future drinking water source quality. The results from this project will be used to improve water quality monitoring in a way that minimizes future societal costs.

Importance for the sector

The results of this project are of importance for a wide variety of entities working in different areas of the water sector. The increasing abundance of low concentrations of organic micropollutants throughout the water cycle is of increasing concern. The tools and guidelines that will be developed in this project are important for all drinking water practitioners. Procedures for the assessment of natural attenuation capacity are essential to understand the risk of micropollutants in the environment. The anticipated results on degradation capacity and rate predications can be directly translated to improved risk assessment of micropollutants present in drinking water resources.

In addition, this project's outcomes can be used in other sectors as well. Tools for assessment of degradation capacity can also be implemented at waste water treatment plants in order to understand and predict the removal of micropollutants in those systems. Additionally, predictions on the residence times required for natural attenuation of micropollutants under specific environmental conditions is necessary in the development of new treatment technologies. By developing assessment tools and guidelines for natural attenuation, this project contributes greatly to an understanding of the presence and improved removal of micropollutants throughout the water cycle.

3. Problem and Goals (max 3 pages)

Problem Background

The increasing analytical capacity to quantify the presence of organic compounds in water resources at low concentrations has led to concern about their presence in the environment. In this project, focus is placed on the presence of these compounds in drinking water resources. Specifically, research will focus on both groundwater and surface water intakes of drinking water. A brief explanation of the water bodies, monitoring, and risk assessment is given below.

Groundwater monitoring: In general, groundwater quality is quite temporally stable. Past and current agricultural and urban activities can pose a threat to quality. The sensitivity of the groundwater to these threats depends on hydrogeological factors which can protect extraction wells. At Vitens, Waterbedrijf Groningen (WBG) and Waterleidingmaatschappij Drenthe (WMD), groundwater recharge areas are identified as being either vulnerable catchments (aquifers with little or no protection by subsurface layers) or not vulnerable catchments (deeper aquifers either partially or fully protected by subsurface layers).

Monitoring of the vulnerable catchments is primarily focused on diffuse contamination with organic micropollutants. Measurements are performed both on the source and in upstream monitoring wells in order to identify future threats. Using groundwater models, monitoring wells are strategically placed at a distance of 10-15 years traveling time from the source to the water intake as a warning system. Source and monitoring wells are sampled infrequently (every 6-8 years) and examined for possible contamination with an extensive analytics package (see Appendix 1). If contamination is found, further monitoring is performed and additional parameters are measured in order to verify the contamination and locate the source. Definitively contaminated wells are monitored more frequently, once a year for source wells or once every other year for monitoring wells. For Vitens, WMD and Waterbedrijf Groningen, the quality of groundwater is increasingly becoming more of an issue, with several treatment plants being interesting test cases for this study (Haarlo/Eibergen in "De Achterhoek", Noordbargeres, and Valtherbos in Drenthe).

Surface water monitoring: WBG also extracts surface water at De Punt (Glimmen) from the river De Drentsche Aa. In contrast to groundwater, there is no physical barrier to protect surface water, and upstream monitoring only predicts quality in the very short term. Therefore, more regular and intensive monitoring of surface water is required.

Surface water samples are collected hourly and combined as a weekly bulk sample. As agriculture is the main source of contamination, these weekly samples are analyzed for a wide range of pesticides. Twice a year, more extensive screening of organic micropollutants is performed, including contaminant groups such as pharmaceuticals, hormones, flame retardants, personal care products, etc. Finally, the toxicity of the surface water is continuously monitored with an online biomonitor (*Daphnia*).

Risk assessment: Current practice at Vitens, WMD, WBG, and Waterlaboratorium Noord (WLN) relies heavily on monitoring to ensure source water quality. Screening for and following micropollutants is performed through sampling and analysis. Once contamination has been established, experts estimate the risks for current and future drinking water sources. In such estimates, the natural attenuation capacity of the subsurface is taken into account globally, mainly based on literature data. However, the lack of literature data for organic micropollutants makes it difficult to predict natural attenuation. Often, either biodegradation rates of these new compounds have not yet been determined or the environmental conditions used for published degradation tests differ from those encountered at locations throughout the Netherlands. This results in an unpredictable variable margin of error in the risk assessment. To reduce the errors in current risk assessment, location specific natural degradation potential must be properly estimated. Through integrating site specific measurements with degradation potential, this project aims to improve understanding of natural attenuation capacity and thus improve model estimates.

<u>Goals</u>

This project aims to increase the understanding of the natural attenuation capacity for degradation of organic micropollutants in order to improve the prediction capacity of models. The application of known assays and development of new molecular tools will give insight into the degradation capacity present. Integration of molecular data with information on environmental conditions at locations with and without degradation capacity will be used for guidelines which predict the expected degradation at a particular location. This project is split into four tasks; specific goals and results for each task are given in section 5 (Execution of the project) below. An overview of the overall results are:

- Analytical capacity to measure degradation genes of currently known and yet undiscovered microbes
- Screening of in situ degradation capacity and microbial diversity at locations investigated within this study
- Determination of degradation capacity and degradation rates ex situ under controlled microcosm conditions
- Guidelines on predicting natural attenuation capacity for the compounds investigated
- 2-3 scientific publications

The outcomes of this project are an important step in improving the understanding of natural attenuation of micropollutants in water cycles. Following this project, the results will be directly applied to improve risk assessment models used to understand the quality of drinking water resources and predict future security of intakes. Additionally, as described in section (2), these results can contribute to other sectors in the water cycle where micropollutants are a threat.

4. Collaborators (1-2 pages)

Partners

Department of Environmental Technology, Wageningen University Wageningen, The Netherlands

www.ete.wur.nl

The department of Environmental Technology (ETE) has nearly fifty years of experience research on remediation of environmental pollution problems using natural attenuation processes. Previous and current work focuses on the presence of pollutants in surface, ground, and waste water systems and the use of biological processes to remove and valorise resource streams. Under the guidance of Prof. Huub Rijnaarts, increasing focus is being placed on the removal and environmental risk of micropollutants. To this end, TT Assistant Professor Dr. Alette Langenhoff leads a number of PhD projects on biodegradation processes in water treatment techniques for the removal of organic micropollutants. This work is strengthened by Prof. Tinka Murk's research assessing the eco-toxicology of organic pollutants. Finally, ETE has a strong background in collaboration with private sector entities to ensure that fundamental research is translated into applicable technologies. Thus, this proposed project fits well within the scientific background and research capabilities of ETE.

Laboratory of Microbiology, Wageningen University Wageningen, The Netherlands

www.mib.wur.nl

The Laboratory of Microbiology (MIB) is engaged in research and education on biotransformations and interactions of microorganisms as well as their control. Moreover, it contributes to the exploitation of the generated knowledge in the application areas of Health & Food, Bioproducts & Energy, and Environment & Sustainability. The research is primarily molecule-driven and genomics-based while incorporating systems approaches at all levels. Within MIB, the Molecular Ecology Group, headed by Prof. Hauke Smidt, focuses on the integrated application of innovative cultivation and high throughput as well as dedicated biomolecular approaches to understand microbial ecology in the environment. To this end, previous and current projects aim at identifying degraders of organic contaminants, illuminating degradation pathways, understanding the environmental conditions and symbiotic relationships required, and developing molecular assays to detect and quantify biomarkers predictive of ecosystem diversity and functioning. The expertise and facilities at MIB on the development and application of molecular tools for environmental samples is integral to the success of the project.

WLN Glimmen, The Netherlands

www.wln.nl

WLN is the water quality and water technology centre of two Dutch Water Companies:

Waterleidingmaatschappij Drenthe and Waterbedrijf Groningen. For these water companies and others (industrial water suppliers, industries, water boards, hospitals etc.), WLN connects (applied) research and every day practice. Dr.ir. Peter van der Maas coordinates strategic research and development on water quality (monitoring and management) and water technology.

Vitens

Leeuwarden, the Netherlands

www.vitens.nl

Vitens is the largest drinking water company in the Netherlands and supplies drinking water in the provinces of Friesland, Overijssel, Gelderland, Flevoland and Utrecht. The core business of Vitens is supplying drinking water to 5.4 million people with a total production of 330 million m³ annually. Vitens serves as both the operator and owner of all physical assets. The physical assets comprise 95 groundwater treatment plants, a distribution network of 47,500 km and 2.4 million metered connections. Dr. ing. Bendert de Graaf is a project coordinator for Vitens and operates out of the laboratory. With a PhD in Molecular and Medical Genetics, and a Masters and Bachelors in Applied Genetics, he is seen as one of the sensor and online water quality monitoring experts at Vitens. Dr. de Graaf works within the Smart Grid management team.

5. Execution of the project (max 3 pages)

Input and rights of partners

The collaboration between research at Wageningen University and private sector drinking water companies is essential to the execution of this project. The fundamental knowledge at ETE and MIB will be used to develop practical assessment tools and guidelines for WLN (WMD and WBG) and Vitens. The inputs of each partner towards the various tasks are described below. In general, WLN and Vitens will provide extensive monitoring of groundwater and surface water intakes for micropollutants as well as geochemical characterization of the water. Additionally, groundwater samples will be provided when required. ETE and MIB will execute research on determining the natural attenuation capacity for degradation of these micropollutants. The interaction will result in tools and guidelines for assessing natural attenuation capacity. The rights to these results are described in the collaboration contract ("sammenwerkingsovereenkomst").

Execution of tasks

Phase 1: Selection of compounds and locations

First, a number of compounds will be selected for research. Existing monitoring data from measurements of groundwater and surface water locations will be interpreted. Data on temporal and spatial changes in contaminant concentrations will be used to give a first estimate of locations, environmental characteristics, and other conditions that are promising in terms of natural attenuation. Additionally, previous monitoring data will be used to identify a list of compounds that will be focused on during this research. For this, the overall threat of the compound will be the most important factor; however, the chances for natural attenuation and the availability of other literature on biodegradation will be taken into consideration. In addition to pesticides, which are currently seen as the largest threat, the list will most likely include pharmaceuticals observed in surface water systems. Phase 1 will result in: (a) a list of interesting locations for further investigation, (b) a list of specific compounds to be researched, (c) a literature review for these compounds, especially with a list of known degraders, degradation rates, degradation genes, and degradation conditions.

Phase 2: Biodegradation assessment with existing molecular tools

The natural degradation potential will be assessed using established molecular tools for the list of compounds and locations identified in phase 1, both in situ and ex situ. For both approaches, (ground)water samples will be required from field locations. This work will rely on the application of existing molecular assays to determine the presence and abundance of a functional degradation gene (qPCR) or a specific degrader (phylogenetic analysis based on next generation sequencing of 16S rRNA genes). Degradation capacity will be estimated in situ by analyzing field-collected groundwater samples using the aforementioned molecular tools. Ex situ capacity will be investigated with degradation experiments using field collected water as an inoculum. In these microcosms, degradation capacity can be enriched under controlled conditions with amendments to enhance microbial growth. In addition to measurements of compound concentrations to determine degradation rates, above-mentioned molecular tools will be applied using DNA and RNA extracted from the microcosms to assess enrichment and activity of specific microorganisms and their degradative pathways. This work will be integrated into current PhD projects focusing on degradation experiments with micropollutants. Phase 2 will result in an assessment of the known degradation capacity and degradation conditions at locations both in situ and under enhanced conditions ex situ.

Phase 3: Development of new molecular tools for biodegradation assessment

New tools will be developed for the determination of degradation capacity. Whereas phase 2 focused on the detection of known degraders and functional genes, this phase will focus on identifying degraders not previously reported in literature because the compound or the specific environmental condition has yet to be investigated. To some extent, a similar setup of in situ and ex situ work will be performed and groundwater samples will once again be required. Assessment of phylogenetic diversity using next generation sequencing of field samples will give insight into microbial communities associated with contamination. Additionally, microbial community analysis of ex situ degradation experiments will give additional information on the identity of potential degraders and associated degradation. Additionally, further investigation will be performed to determine potential genes essential to the degradation and develop new molecular tools for their measurement. Finally, degradation experiments will be utilized from other PhD projects, thereby expanding the dataset. Overall, phase 3 will result in new data on degraders and degradation genes to fill literature gaps in current data.

Phase 4: Setting guidelines for natural attenuation capacity prediction using molecular tools

All data will be integrated into guidelines for prediction of the natural attenuation capacity. In this phase, results from phase 2 and 3 on the presence of degradation capacity, environmental conditions required for degradation, and degradation rates will be integrated. At this stage, additional groundwater sampling and analysis will be performed at locations not previously investigated where degradation is expected. The results will be used to further confirm the newly developed tools from phase 3 and verify prediction guidelines. Phase 4 will result in a set of guidelines for the use of molecular tools combined with environmental characteristics that can be used to predict natural attenuation of the compounds identified in phase 1.

Time-plan

	Time (in months)					
Task	1-6	7-12	13-18	19-24	25-30	31-36
Phase 1						
 Analysis of current monitoring data 						
 Identification of key compounds 						
 Identification of key locations 						
Literature review						
 Integration with current PhD projects 						
Phase 2						
 Setting up qPCR protocols 						
 Degradation experiments 						
 Molecular analysis of in situ and ex situ 						
samples for known degraders						
Phase 3						
 Degradation experiments 						
Molecular analysis for in situ and ex situ						
samples to find new degraders						
Development of new molecular tools						
Phase 4						
 Integration of data 						
 Development of guidelines 						
 Application to other locations 						

6. Budget

Subsidy		Total Project	
	1 year	(3 years)	
TKI subsidy (2013 estimate)	83498	166996	
5% administration costs		-8350	
Actual TKI subsidy	79323	158646	

Project Budget		Total Project	
	1-year	(3 years)	
Salary 3 year post-doc (0.50 fte)	57882	173646	
Salary 1 year PhD (0.5 fte)	33792	33792	
Consumables	26667	80000	
Travel costs	5000	15000	
Total Cash	123341	302438	
WLN			
in-kind analyses, sampling, etc.	50406	151219	
Vitens			
in-kind analyses, sampling, etc.	50406	151219	
WUR			
Research support	20163	60488	
Total in kind	120975	362926	
Total project costs	332682	665364	

7. Knowledge sharing and dissemination

TKI Watertechnologie is welcome to publish the names and contact details of the project participants, the project title, and a short summary of the project on the Topsector Water website. The public summary can be found below.

Author: Nora B. Sutton
 Partners: Department of Environmental Technology, Wageningen University

 Laboratory of Microbiology, Wageningen University
 Water Laboratorium Noord
 Vitens

 Title: MicroNAC - Advanced tools for assessment of the Microbiological Natural Attenuation Capacity for Micropollutants in the water cycle
 Topsector: Water
 Start: December 2013
 End: December 2016

English Summary

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This project fits within the Topsector Water's focus on "Water for All" by developing sustainable, cost-effective practices to ensure the future and water quality of Dutch drinking water resources. By establishing tools to assess natural attenuation capacity and degradation rates, this information can be incorporated into risk assessment models used to predict the future security of drinking water resources. The economic and social benefits of clean source drinking water practices are numerous.

Nederlandse Samenvatting

De toenemende aanwezigheid van organische microverontreinigingen in verschillende onderdelen van de watercyclus kunnen in de nabije toekomst een bedreiging vormen voor waterreserves. Lage concentraties van deze verbindingen worden momenteel gedetecteerd in grond- en oppervlaktewater dat wordt gebruikt als

drinkwater. Hoewel de huidige monitoringstechnieken (chemische analyses) van deze microverontreinigingen, is er weinig bekend over de natuurlijke afbraak van microverontreinigingen onder in situ-omstandigheden. Er is een gebrek aan informatie over de biologische afbraak onder natuurlijke condities en methodes om de natuurlijke afbraak capaciteit te beoordelen zijn noodzakelijk. Deze informatie is nodig om het voorspellende vermogen van bestaande modellen te verbeteren, die gebruikt worden om het lange termijn risico van de verontreiniging van drinkwaterwinningsputten te beoordelen.

Dit onderzoek is gericht op het verbeteren van onze kennis over biologische afbraak van microverontreinigingen. Hiertoe worden instrumenten ontwikkeld om de biologische afbraakcapaciteit van een aantal microverontreinigingen te beoordelen, wat resulteert in beoordelings- en voorspellingsrichtlijnen voor natuurlijke afbraak. Het project zal zich richten op microverontreinigingen die een bedreiging vormen voor de kwaliteit van het Nederlandse drinkwater. Voor deze verbindingen zullen moleculaire technieken op basis van DNA-analyse worden gebruikt om de natuurlijke afbraak capaciteit in het veld te beoordelen. Daarnaast zullen ex-situ afbraak experimenten met veld monsters worden gebruikt om de afbraaksnelheden van bepaalde microverontreinigingen te bepalen. De resultaten zullen worden geïntegreerd om richtlijnen vast te stellen voor de voorspelling van de natuurlijke afbraak met behulp van moleculaire technieken.

Het project zal worden uitgevoerd als een samenwerking tussen private drinkwaterbedrijven en de Universiteit Wageningen. De drinkwaterbedrijven Vitens, WMD en Waterbedrijf Groningen (de laatste twee vertegenwoordigd door Water Laboratorium Noord) zullen expertise bijdragen over de praktijk en de huidige risico's voor drinkwater alsmede monitoringsresultaten en grond- en oppervlaktewater monsters. Binnen Wageningen University zal het werk worden uitgevoerd bij de sectie Milieutechnologie en het Laboratorium voor Microbiologie. Het onderzoek zal zich richten op het ontwikkelen en uitvoeren van instrumenten die gebruikt kunnen worden om de in-situ natuurlijke afbraak capaciteit van specifieke microverontreinigingen te beoordelen. De combinatie van fundamenteel onderzoek op universitair niveau en praktische kennis en expertise geleverd door drinkwaterbedrijven, zal worden gebruikt om richtlijnen vast te stellen voor de beoordeling van het in situ natuurlijke afbraak vermogen van microverontreinigingen.

Dit project past binnen de focus van de 'Topsector Water; water for all' in de ontwikkeling van veilige, duurzame, en efficiënte oplossingen om in de toekomst de waterkwaliteit van Nederlandse drinkwaterbronnen te garanderen. Door de ontwikkeling van instrumenten om de natuurlijke afbraak capaciteit en afbraaksnelheden te beoordelen, kan deze informatie in risicobeoordelingsmodellen worden gebruikt om de toekomstige veiligheid van drinkwaterbronnen te kunnen voorspellen. De economische en sociale voordelen van schone drinkwaterbronnen praktijken zijn talrijk.

Volatile Organ	nic Compounds	Pesticides			Organic Micropollutants	
n-pentaan	1,1-dichloorpropaan	fenol	2,3,6-trichloorfenol	3,4-dimethylaniline	aldicarbsulfon	metoprolol (geen Q)
n-hexaan	1,2-dichloorpropaan	2-methylfenol	2,4,5-trichloorfenol	2,4,6-trimethylaniline	aldicarbsulfoxide	caffeine (geen Q)
n-heptaan	1,3-dichloorpropaan	3+4-methylfenol	2,4,6-trichloorfenol	N-methylaniline	butocarboxim	propranolol (geen Q)
n-octaan	2,2-dichloorpropaan	2,3-dimethylfenol	3,4,5-trichloorfenol	N,N-dimethylaniline	butocarboximsulfoxide	sotalol (geen Q)
n-nonaan	1,1-dichloorpropeen	2,4+2,5-dimethylfenol	2,3,4,5+2,3,4,6-tetrachloor	enolN-ethylaniline	butoxycarboxim	lidocaine (geen Q)
n-decaan	cis 1,3-dichloorpropeen	2,6-dimethylfenol	2,3,5,6-tetrachloorfenol	N,N-diethylaniline	carbetamide	2,4-dinitrofenol
n-undecaan	trans 1,3-dichloorpropeen	3,4-dimethylfenol	pentachloorfenol	2,6-diethylaniline	carbofuran	2,5-dinitrofenol
n-dodecaan	2,3-dichloorpropeen	2-nitrofenol	4-chloor-2-methylfenol	4-isopropylaniline	molinaat	2,6-dinitrofenol
n-tridecaan	1,2,2-trichloorpropaan	2-ethylfenol	4-chloor-3-methylfenol	dibenzylamine	oxamyl	3,4-dinitrofenol
n-tetradecaan	1,2,3-trichloorpropaan	3+4-ethyl+3,5-dimethylfenol	2-chloorfenol	tribenzylamine	propamocarb	D.N.O.C
n-pentadecaan	methylisothiocyanaat	3-nitrofenol	dichloorvos	2-chlooraniline	aldicarb	dinoseb
n-hexadecaan	1,1,2-trichloorpropaan	4-nitrofenol	cis+trans-mevinphos	3+4-chlooraniline	carbendazim	dinoterb
Acetaldehyde	1,4-dioxaan	BDE-028	ethoprofos	2,3-dichlooraniline	joodpropynylcarbamaat	3+4-nitrofenol
benzeen	butylmethylether	BDE-047	sulfotep	2,4+2,5-dichlooraniline	3-hydroxycarbofuran	2,3-dinitrofenol
tolueen	ETBE	BDE-066	atrazine	2,6-dichlooraniline	carbaryl	2-nitrofenol (geen Q)
ethylbenzeen	MTBE	BDE-085	phoraat	3,4-dichlooraniline	chloorpropham	bifenox
n-propylbenzeen	tetrahydrothiofeen	BDE-099	terbutylazine	3,5-dichlooraniline	methiocarb	bromacil
iso-propylbenzeen	tetrahydrofuraan	BDE-100	diazinon	2,3,4-trichlooraniline	pirimicarb	chloridazon
n-butylbenzeen	TAME (geen Q)	BDE-138	desmetryn	2,4,5-trichlooraniline	profam	dimethoaat
iso-butylbenzeen	diisopropylether (geen Q)	BDE-153	disulfoton	2,4,6-trichlooraniline	propoxur	thiabendazool
secundair-butylbenzeen	trichloormethaan (chloroform)	BDE-154	metribucin	3,4,5-trichlooraniline	prosulfocarb	chloridazon-desfenyl
tertiair-butylbenzeen	tetrachloorkoolstof (tetra)	aminocarb	parathion-methyl	2,3,4,5-tetrachlooraniline	swep	chloridazon-methyl-desfenyl
n-pentylbenzeen	broomchloormethaan	barban	fenchloorphos	2,3,5,6-tetrachlooraniline	aminocarb	pendimethalin
o-xyleen	broomdichloormethaan	carbaryl	paraoxon-ethyl	3-chloor-4-methoxyaniline	barban	pyriproxyfen
m+p-xyleen	broomtrichloormethaan	carbofuran	terbutryn	3-chloor-4-methylaniline	dicamba	chloorthalonil
4-isopropyltolueen	dibroomchloormethaan	chloorpropham	triadimefon	4+5-chloor-2-methylaniline	4-CPA	cymoxanil
1,2,3-trimethylbenzeen	tribroommethaan (bromoform)	pirimicarb	parathion-ethyl	3,3'-dichloorbenzydine	2,4-D	epoxyconazool
1,2,4-trimethylbenzeen	1,2-dichloorethaan	profam	bromophos-methyl	4-broomaniline	МСРА	fenmedifam
1,3,5-trimethylbenzeen	1,1,1-trichloorethaan	swep	bromophos-ethyl	o-anisidine	2,4-DP (dichloorprop)	haloxyfop-methyl
1,2,3,4-tetramethylbenzeen	1,1,2-trichloorethaan	propoxur	methidathion	2-nitroaniline	МСРР	imidacloprid

Appendix 1. Compounds monitored in this study.

1,2,3,5-tetramethylbenzeen	1,1,2,2-tetrachloorethaan	3-hydroxycarbofuran	ethion	3-nitroaniline	2,4,5-T	lenacil
1,2,4,5-tetramethylbenzeen	trichlooretheen (tri)	methiocarb	azinphos-methyl	4-methoxy-2-nitroaniline	2,4-DB	metamitron
2-ethyltolueen	tetrachlooretheen (per)	prosulfocarb	pyrazophos	4-methyl-2-nitroaniline	МСРВ	dimethenamid
3-ethyltolueen	hexachloorethaan	a-HCH	etrimphos	4-methyl-3-nitroaniline	2,4,5-TP	tebuconazool
4-ethyltolueen	hexachloorbutadieen	b-HCH	fenitrothion	2,6-dichloor-4-nitroaniline	bentazon	thiacloprid
1,2-diethylbenzeen	1,1,1,2-tetrachloorethaan	d-HCH	malathion	2-phenylsulfonaniline	fluroxypyr	1,2-benzisothiazool-3-on
1,3-diethylbenzeen	1,2-dibroomethaan	g-HCH (lindaan)	cis-chloorfenvinphos	pentachlooraniline	aminomethylfosfonzuur	boscalid
1,4-diethylbenzeen	1,2-dibroom-3-chloorpropaan	НСВ	propazine	diflufenican (geen Q)	glyfosaat	bromoxynil
1,3-diisopropylbenzeen	1-broom-3-chloorpropaan	PCNB	trietazine	epoxyconazool	chloortoluron	dimethomorf
1,3,5-triisopropylbenzeen	dibroommethaan	dichlobenil	fonofos	esvenvaleraat	diuron	dithianon
styreen	epichloorhydrine	aldrin	prometryn	ethofumesaat	isoproturon	fluazinam
naftaleen	1-chloorpentaan	dieldrin	bromacil	fenamifos	linuron	fluopicolide
biphenyl	chloormethaan	endrin	cyanazine	fenpropimorf	metabenzthiazuron	flutolanil
biphenylether	dichloormethaan	heptachloor	trans-chloorfenvinphos	fluazifop-butyl	metoxuron	irgarol
chloorbenzeen	chlooretheen (vinylchloride)	cis-heptachloorepoxide	tetrachloorvinphos	folpet (geen Q)	monolinuron	lambda-cyhalothrin
1,2-dichloorbenzeen	1,1-dichlooretheen	trans-heptachloorepoxide	simazine	kresoxim-methyl	monuron	mesosulfuron-methyl
1,3-dichloorbenzeen	cis-1,2-dichlooretheen	a-endosulfan	benazolin-ethylester	metalaxyl	metobromuron	methomyl
1,4-dichloorbenzeen	trans-1,2-dichlooretheen	b-endosulfan	ametryn	fosalon	chloorbromuron	quizalofop-p-ethyl
1,2,3-trichloorbenzeen	chloorethaan	op-DDE	atrazine-desethyl	fosfamidon-a	fenuron	triallaat
1,2,4-trichloorbenzeen	1,1-dichloorethaan	pp-DDE	atrazine-desisopropyl	fosfamidon-b	chlooroxuron	triflusulfuron-methyl (geen
1,3,5-trichloorbenzeen	3-chloor-1-propeen	op-DDD	trichloronat	propiconazool-a	fluometuron	difenoconazool (geen Q)
2-chloortolueen	2-chloor-1-propeen	pp-DDD	azinphos-ethyl	propiconazool-b	pencycuron	paclobutrazool (geen Q)
4-chloortolueen	cis 1-chloor-1-propeen	op-DDT	chlorpyrifos-ethyl	thiabendazool	diglyme	cinidon-ethyl
cyclohexaan	trans 1-chloor-1-propeen	pp-DDT	chlorpyrifos-methyl	oxadixyl	triglyme	cloquintocet-mexyl
methylcyclohexaan	dichloordifluormethaan	pentachloorbenzeen	demeton-S-methyl	etridiazool (geen Q)	tetraglyme	cypermethrin
cyclohexeen	trichloorfluormethaan	tecnazeen	dimethoaat	aclonifen (geen Q)	metaldehyde	dodemorf
1,2,3,4-tetrachloorbenzeen	freon-113	propachloor	fenthion	azoxystrobine	aldicarb	endothall
1,2,3,5+1,2,4,5-tetrachloorbenze	en	alachloor	hexazinon	bifenox	molinaat	fenhexamide
broombenzeen		metazachloor	methacrifos	broompropylaat (geen Q)	pendimethalin	florasulam
		cis-permethrin	paraoxon-methyl	carbofenthion	pyriproxyfen	fluxapyroxad
		trans-permethrin	penconazool	chloroneb	carbamazepine	joodsulfuron-methyl
		dichloran	pirimiphos-methyl	deet		metsulfuron-methyl

metolachloor isobenzan (telodrin) mirex methoxychloor pentachlooraniline PCB-28 PCB-52 PCB-101 PCB-118 PCB-138 PCB-138 PCB-153 PCB-153 PCB-153 PCB-180 2,6-dichloorbenzamide (BAM isodrin vinchlozolin cis-chloordbenzamide (BAM isodrin vinchlozolin cis-chloordbenzamide (BAM isodrin vinchlozolin cis-chloorbenzamide (BAM	tolclofos-methyltriazophoschloridazondeltametrintriadimenoltrifluralinprometonnaftaleenacenaftyleenacenaftyleenacenafteenfluoreenfluoratheenpyreenbenzo(b)fluorantheenbenzo(b)fluorantheenbenzo(a)pyreenbenzo(a)pyreenbenzo(b)fluorantheenbenzo(b)fluorantheenbenzo(b)fluorantheenbenzo(b)fluorantheenbenzo(b)fluorantheenbenzo(b)fluorantheenbenzo(a)pyreendibenz(ah)antraceenchryseenbenzo(b)fluorantheenbenzo(b)fluorantheenbenzo(b)fluorantheenbenzo(a)pyreenailineanilineo+m+p-toluidine	2,4,6-tribroomanisol 2,4,6-tribroomfenol 2,4-dibroomfenol 2,6-dibroomfenol 3-broomfenol Heptachloorepoxide (cis + tra	ibuprofen sulfametoxazool atenolol (geen Q) diclofenac (geen Q) ketoprofen (geen Q) naproxen (geen Q) ns) paracetamol (geen Q)	procymidon propyzamide pyroxsulam quinoxyfen thiofanoxsulfoxide tribenuron-methyl triclopyr clopyralid picloram bupirimaat sulcotrion mesotrion nicosulfuron
3,5-dichloorfenol	aniline	hiline		