23-04-2024 TKI SFVI

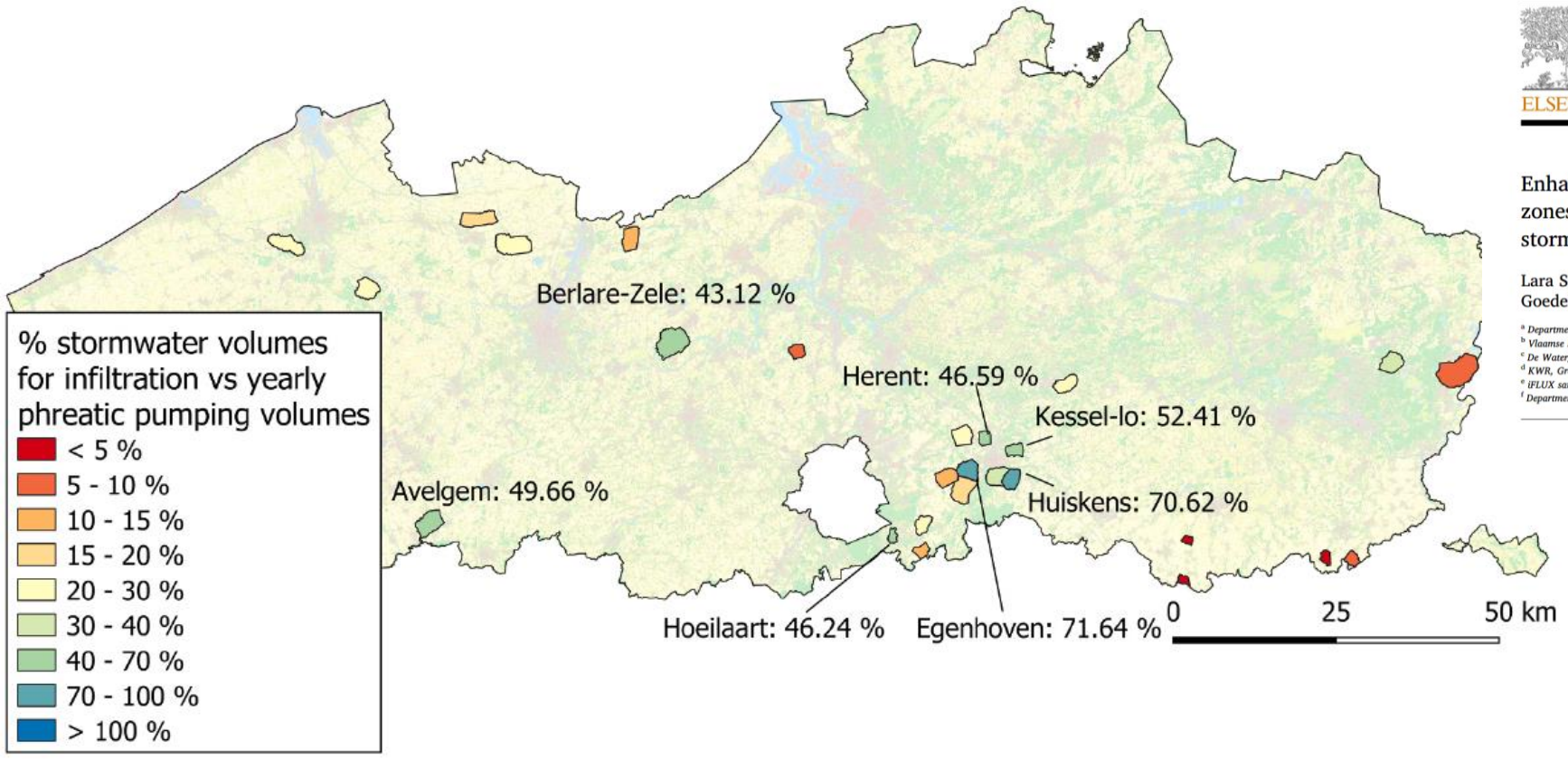
Waterkwaliteit afstromend hemelwater

Wat weten we?

KWR

Bridging Science to Practice

Afkoppelen verhard oppervlak kan significant bijdragen
Vergelijking met freatische drinkwaterwinning in Vlaanderen



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journal homepage: www.elsevier.com/locate/ejrh

Enhancing groundwater recharge in drinking water protection zones in Flanders (Belgium): A novel approach to assess stormwater managed aquifer recharge potential

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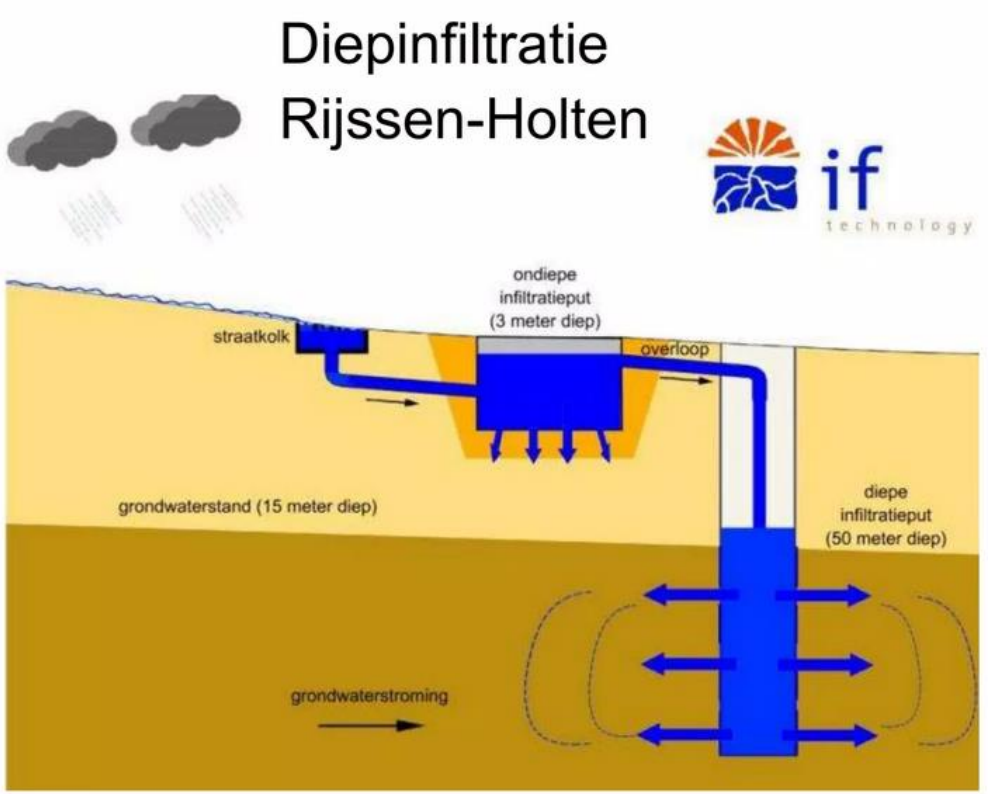
[f] Department of Earth and Environmental Sciences, KU Leuven, Celestijnenlaan 200E, Heverlee 3001, Belgium

Fig. 8. Comparison of stormwater volumes for recharge versus yearly pumping volumes for drinking water production from phreatic aquifers. Protection zones with highest percentages (> 40%) indicated with labels.

Maar is dat water dan wel schoon genoeg?



Wat zit er eigenlijk in afstromend hemelwater?

- 1982: eerste uitgebreide studie in NL
- Daarna groot aantal studies bij gemeenten, waterschappen en Rijk

- 2007: studies samen in één database (Boogaard & Lemmen, 2007)
- 2019: update database (Liefting et al. 2020)
  - 1742 monsters
  - 191 locaties
  - Nutriënten, zware metalen, zouten, minerale olie, PAK, FIB en beperkt aantal pesticiden

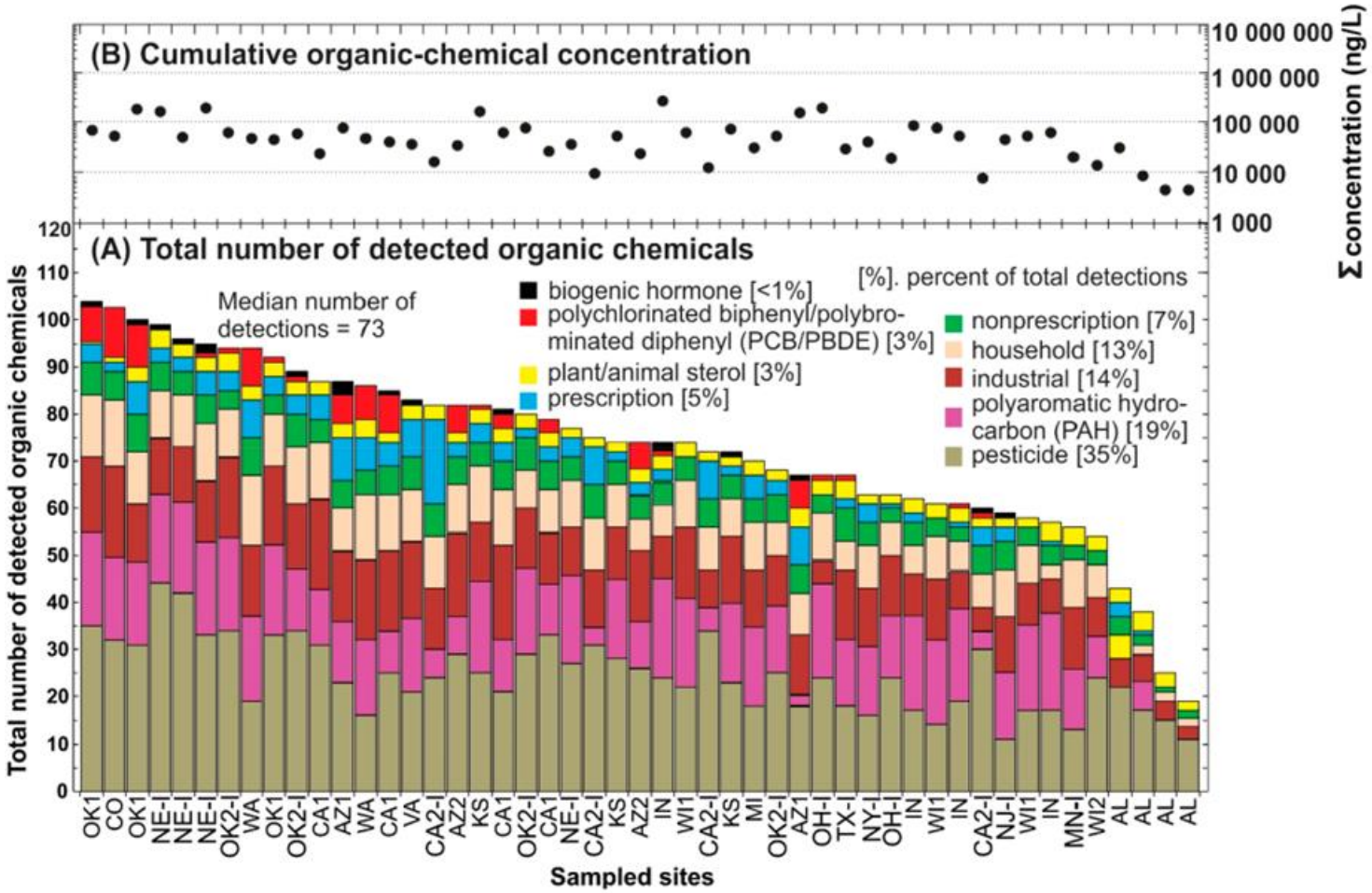
Table 6: Median, d10 and d90 of 16 commonly analyzed substances in the STOWA database on runoff quality (Liefting et al., 2020)

Substance	Unit	Median	d10	d90
Cadmium (Cd)	µg/l	0.09	0.01	0.58
Copper (Cu)	µg/l	11	3.3	41
Mercury (Hg)	µg/l	0.02	<0.02	0.07
Lead (Pb)	µg/l	8	1.8	86
Nickel (Ni)	µg/l	1.9	0.5	8.0
Zinc (Zn)	µg/l	70	11	290
Anthracene	µg/l	0.002	0.001	0.0077
Benzo(a)-pyrene	µg/l	0.004	0.002	0.031
Mineral oil	µg/l	60	<50	470
COD	mg/l	25	12	76
P-total	mg P/l	0.21	0.06	0.52
N-Kjeldahl	mg N/l	1.5	0.68	4.0
Ammonium NH_4-N	mg N/l	0.48	<0.05	2.16
Nitrate NO_3-N	mg N/l	0.72	0.32	2.2
TSS	mg/	14	4.2	70
E coli	cfu/100 ml	$6.2\ 10^2$	$2.1\ 10^1$	$2.1\ 10^4$

Recente buitenlandse studies wijzen op aanwezigheid breed scala organische microverontreinigingen (omv's)

Masoner et al. (2019):

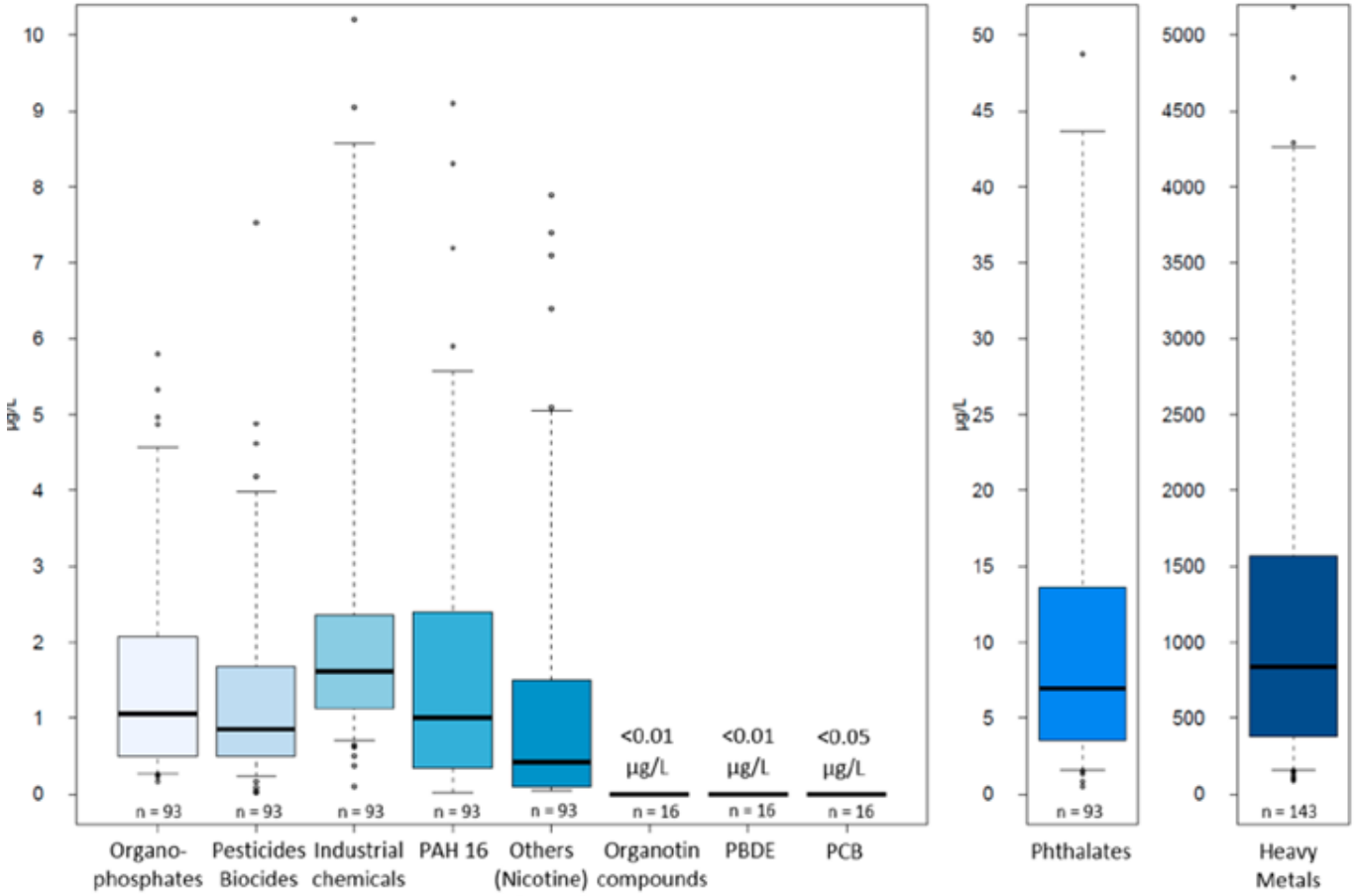
- 52 events, 21 sites in USA
- 438 omv's onderzocht
- 215 omv's aangetroffen
- 73 verschillende stoffen (mediaan)
- Cum. concentratie 4,37 µg/l tot 263 µg/l



Recente buitenlandse studies wijzen op aanwezigheid breed scala organische microverontreinigingen (omv's)

Wicke et al. (2021):

- 5 catchments in **Berlijn**
- 106 stoffen onderzocht
- 71 stoffen aangetroffen
- Gemiddelde cum. concentratie omv's: 24 µg/l
- Ftalaten en zware metalen in hoogste concentraties
- Ftalaten mogelijk afkomstig van de coating onderkant auto's



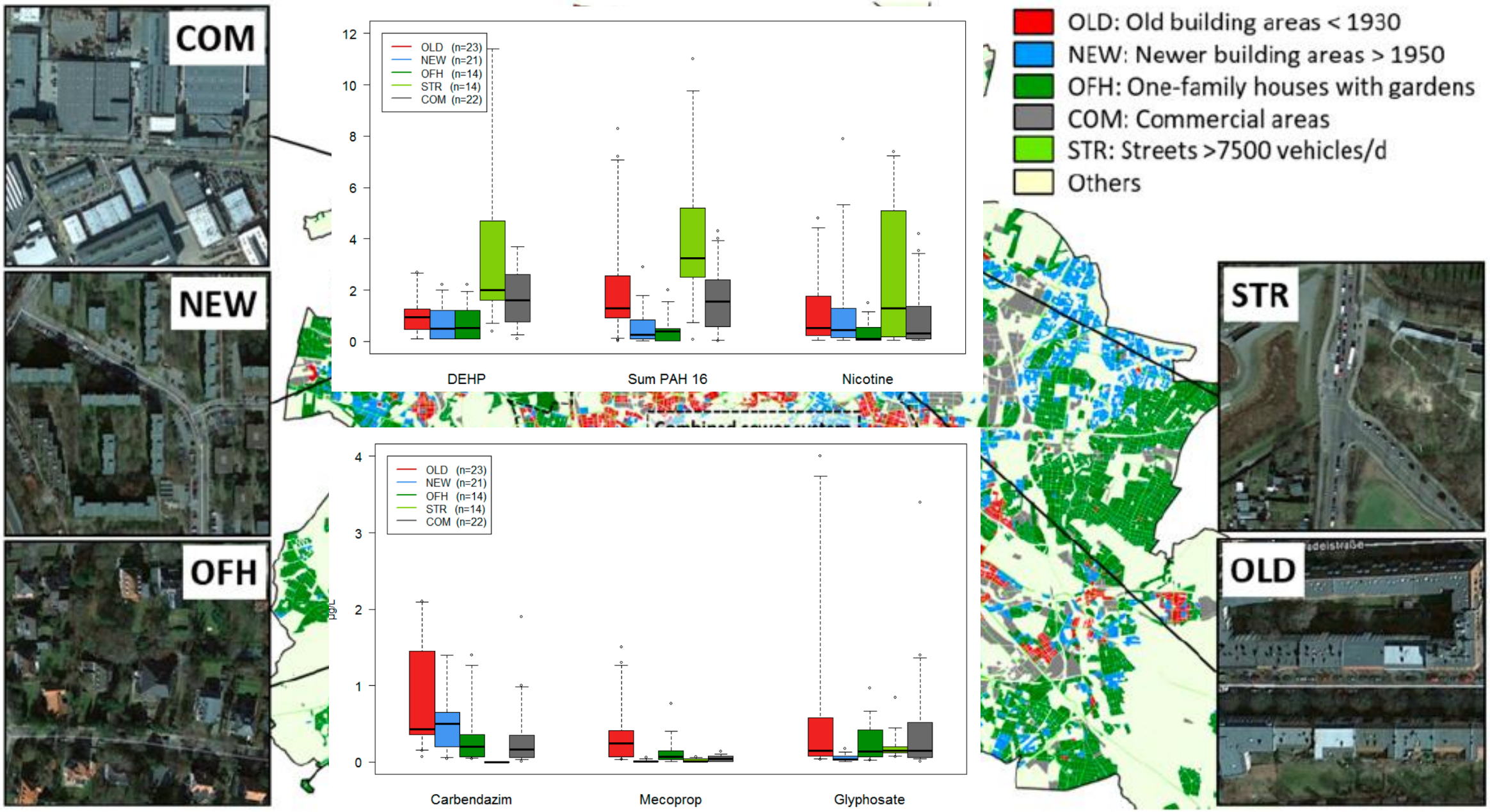


Figure 1. Land-use types and location of monitoring catchments in Berlin (Pictures © Google 2015, Geobasis-DE/BKG).

Recente buitenlandse studies wijzen op aanwezigheid breed scala organische microverontreinigingen (omv's)

Gasperi et al. (2022):

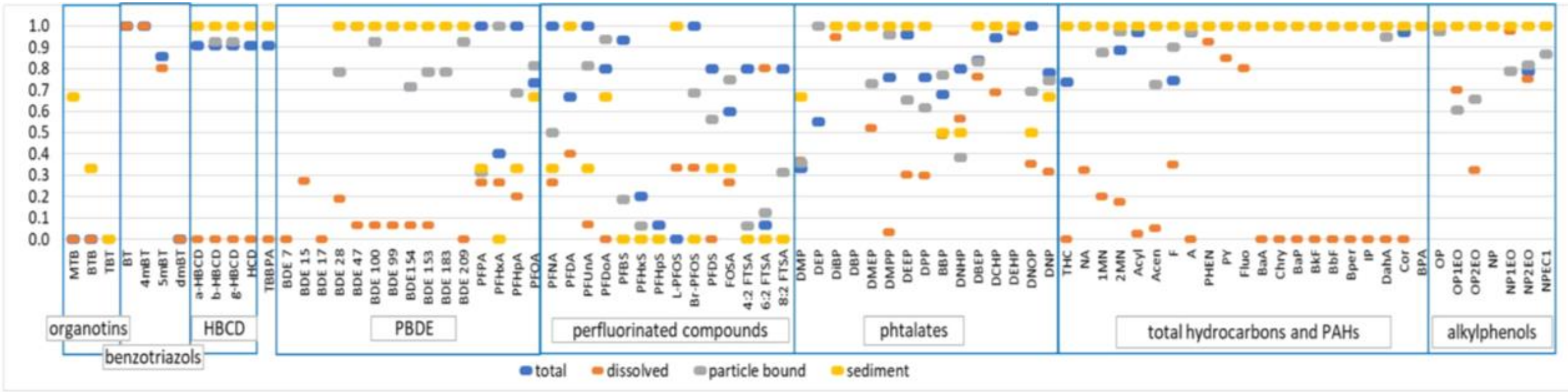
- 5 catchments in **Parijs** gericht op verkeer
- 128 stoffen onderzocht
- Normoverschrijdingen (Europees + Frans):
  - 100 x: benzo(a)pyreen, fluorantheen
  - 10-100 x: Cu, DEHP (Bis(2-ethylhexyl)ftalaat)
  - 1-10 x: As, Sb, Zn, bisphenol A, 4-tert-actylphenol, nonylphenol, polybrominated biphenyl ethers (som), PFOS, Hexabromocyclododecane (som), Anthracene
- PFAS duidelijk aanwezig met concentraties voor individiuele stoffen oplopend tot 125 ng/l
- PFOA dominant gevolgd door 6:2 FTSA, PFOS en andere perfluorcarbonzuren

Is grondwatervervuiling tegen te houden?

Gasperi et al. (2020): poging tot verdeling over verschillende fases:

PFAS: veel minder in sediment, opgelost en in deeltjes

PAK: weinig opgelost, vooral aan sediment



Indicatie voor NL op basis van metingen in RWZI-influent

Langeveld et al. (2020):

- Ontmengen RWZI influent stroom aan de hand van tracers voor de droogweerafvoer (o.a. diclofenac)
- 20 stoffen > 2x detectie limiet
- Herbicides (o.a. glyphosaat, diuron), pesticides (o.a. fipronil, permethrin), fungicides, vlamvertragers (2) en een weekmaker
- Groot aantal bestrijdingsmiddelen in lijn met metingen van Masoner et al. (2019) en Wicke et al., 2021
- **Groot aantal stoffen nog niet in beeld in NL!**

Verkeer een belangrijke bron

- Verbranding brandstoffen (o.a. PAK)
- Slijtage van autobanden en remmen (o.a. 6PPD, Sb, Cu)
- Smeermiddelen, olie en coatings (o.a. PFAS)
- Coatings (weekmakers)

Pollution from car tires is killing off salmon on US west coast, study finds

Mass die-offs of coho salmon just before they are about to spawn have been traced to tire fragments washed into streams by rain

Coho salmon, which can grow to 2ft in length, spend their lives in the ocean but return to the US Pacific coast to spawn. Photograph: NOAA/Alamy

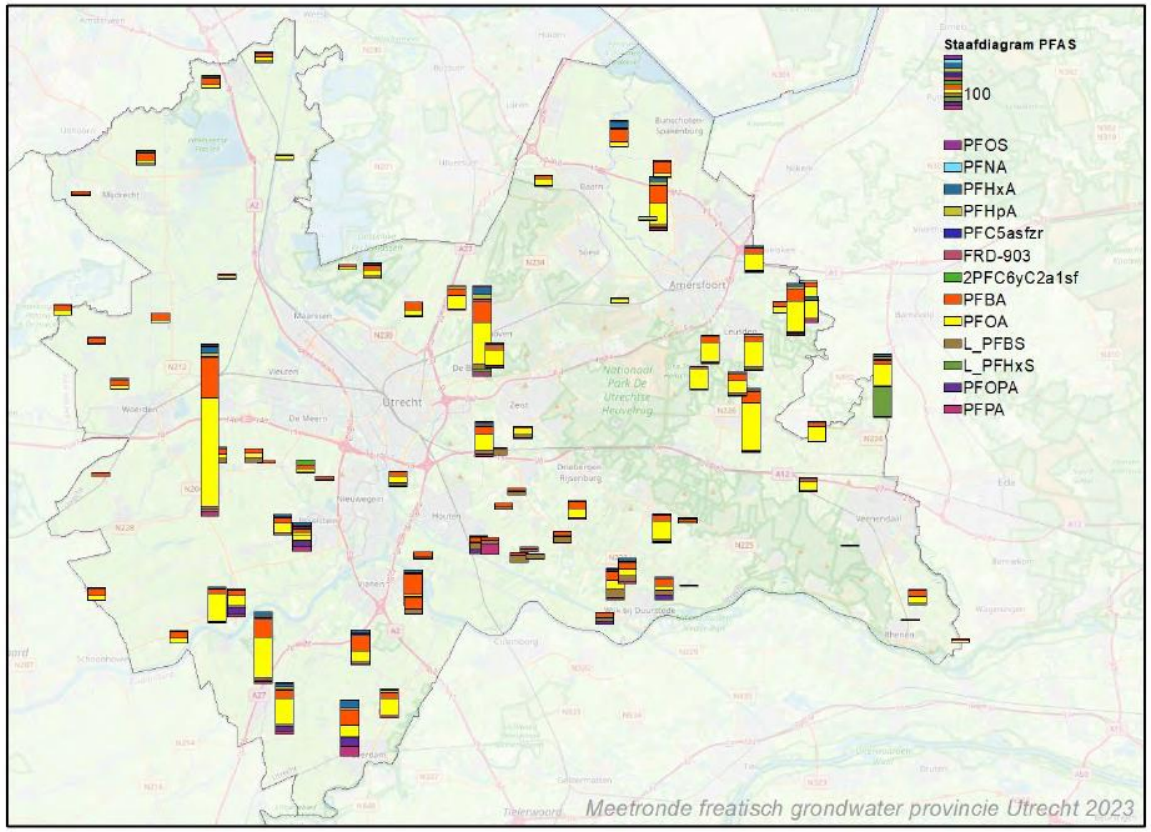
N-(1,3-Dimethylbutyl)-N′-phenyl-p-phenylenediamine-quinone (6PPD-quinone)

Afbraakproduct van 6PPD (antioxidant in rubber)

PFAS……

- Provincie Utrecht freatisch meetnet meetronde 2023: PFOA, PFBA relatief vaak **onder wegbermen** en boven drinkwaternorm

- Zhu & Kannan (2020): meting PFAS in 18 autosmeermiddelen zowel vooraf (71.6 ng/l) als na oxidatie (1840 ng/l)
- Global Automotive Declarable Substance List (GADSL, 2018): PFAS group -> 2953 stoffen



Kaart 3: Beeld van de somconcentratie van PFAS-stoffen in nanogram per liter in de onderhavige meetronde 2023.

Bestrijdingsmiddelen…

- Waarom vinden we niet toegelaten gewasbeschermingsmiddelen (o.a. diuron, fipronil, imidacloprid)?
- Wel toelating voor ander gebruik: biocide in coatings, diergeneesmiddel
- Hoge doseringen bij huisdieren: mg/kg
- Deels ook met fluorverbindingen (o.a. Flurolaner, Fipronil, Afoxolaner)

Vinkocide CDO

Ref: RC024

Substances: Diuron, Carbendazim, OIT

Vinkocide™ CDO is a formulation of Carbendazim, Diuron and 2-n-Octyl-2H-isothiazol-3-one (OIT)

Vinkocide™ CDO is especially suitable for the protection against algal and fungal attack for surface and facade coatings as well as for other building chemical products.
Vinkocide™ CDO is suitable for the preservation of a number of products because of its favorable properties. Experience shows an excellent compatibility with most products, but we recommend testing the compatibility in individual formulations.

Product Benefits

- For the preservation of dry paint films and coatings for outdoor applications
- Excellent stability and long-termefficacy
- Free from heavy metals, organic solvents and VOC
- Compatible with nearly all raw materials for water-basedproducts
- Free from heavy metals, organicsolvents and VOC

Product Types

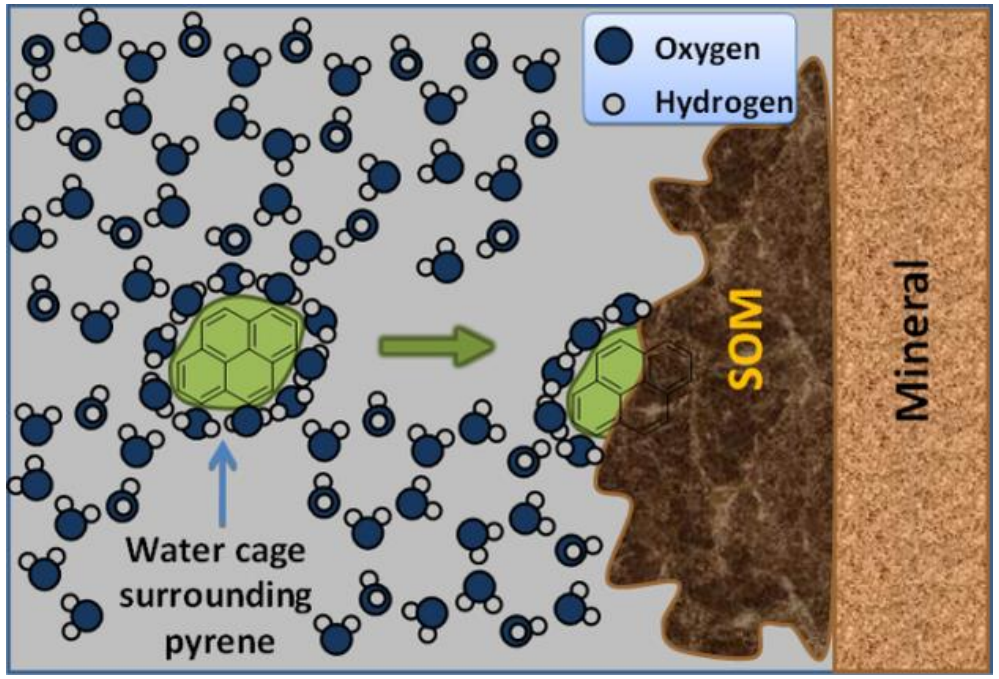
- Film preservatives

Bestrijdingsmiddelen…

- Waarom vinden we niet toegelaten gewasbeschermingsmiddelen (o.a. diuron, fipronil, imidacloprid)?
- Wel toelating voor ander gebruik: biocide in coatings, diergeneesmiddel
- Hoge doseringen bij huisdieren: mg/kg
- Deels ook met fluorverbindingen (o.a. Flurolaner, Fipronil, Afoxolaner)

Verwijdering van organische microverontreinigingen tijdens bodempassage

Sorptie



Afbraak

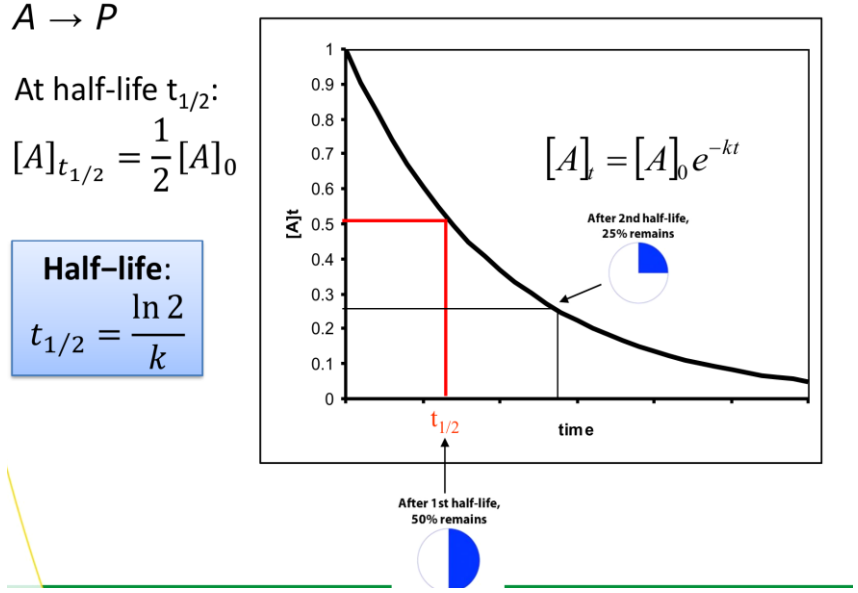
$A \rightarrow P$

At half-life $t_{1/2}$:

$$[A]_{t_{1/2}} = \frac{1}{2}[A]_0$$

Half–life:

$$t_{1/2} = \frac{\ln 2}{k}$$



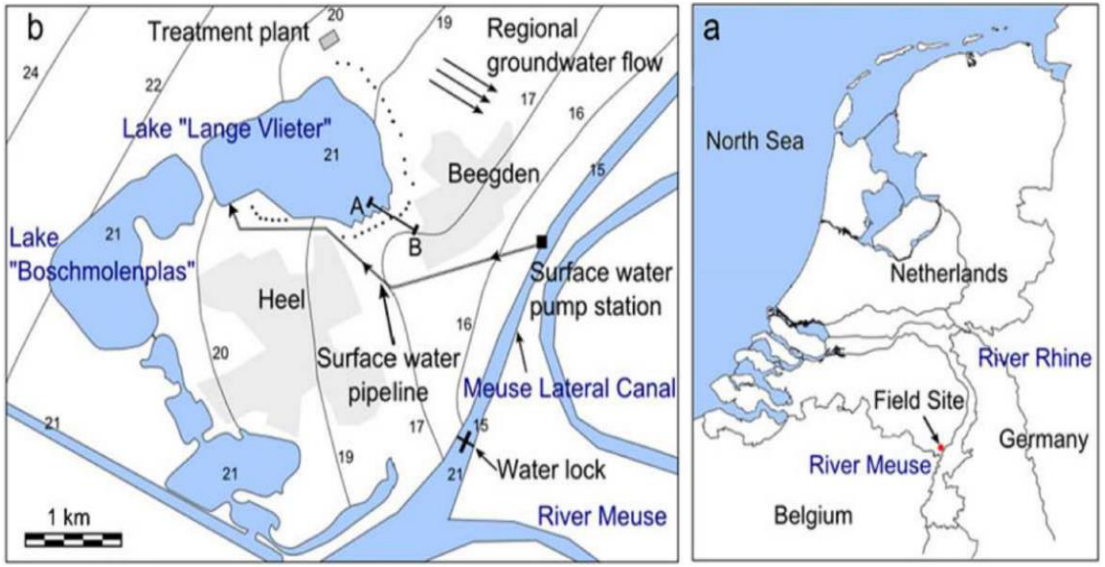
Verwijdering van organische microverontreinigingen tijdens bodempassage

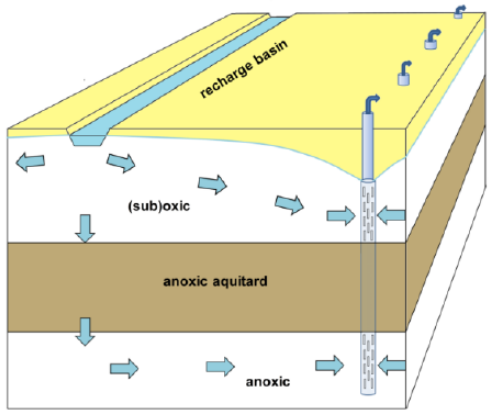
Kennis vanuit de drinkwatersector

duineninfiltratie

oeverinfiltratie

Oeverinfiltratie – waterproductiebedrijf Heel

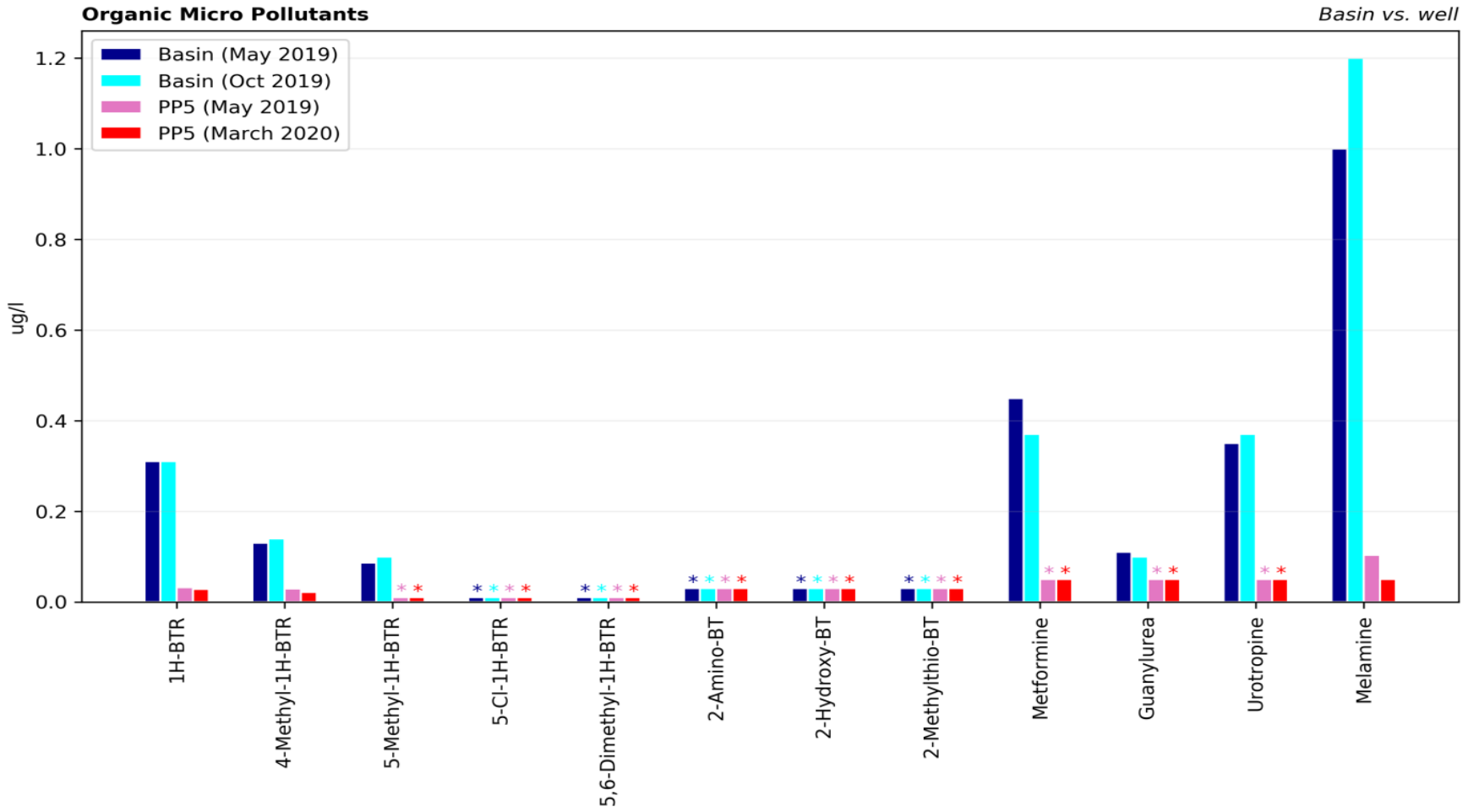




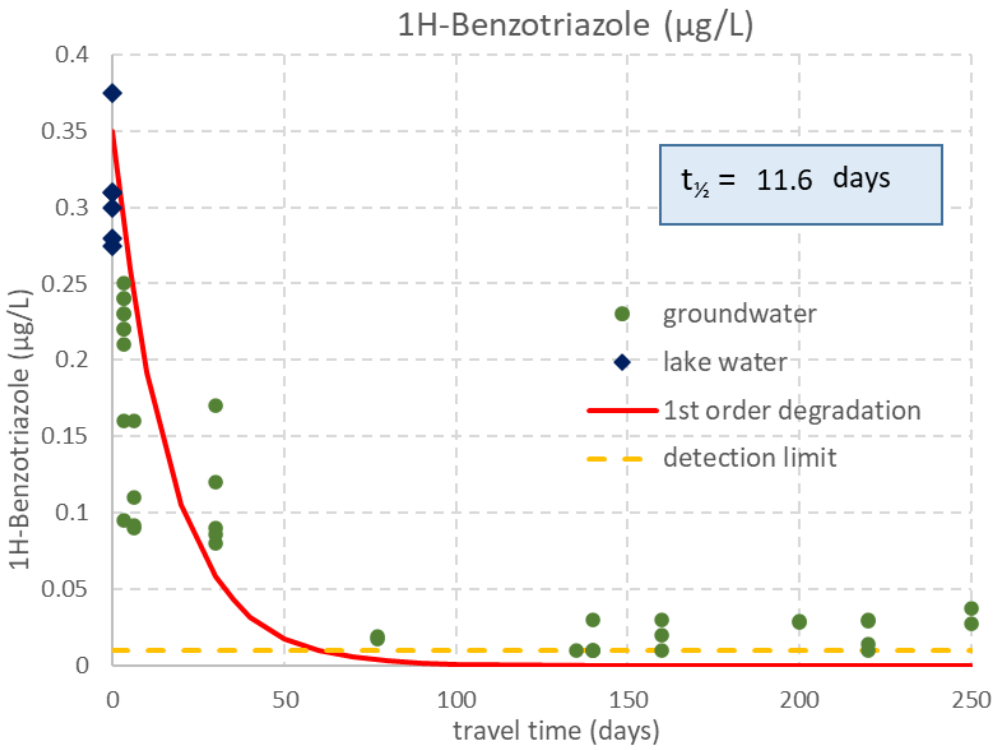
spaarbekken'De Lange Vlieter'

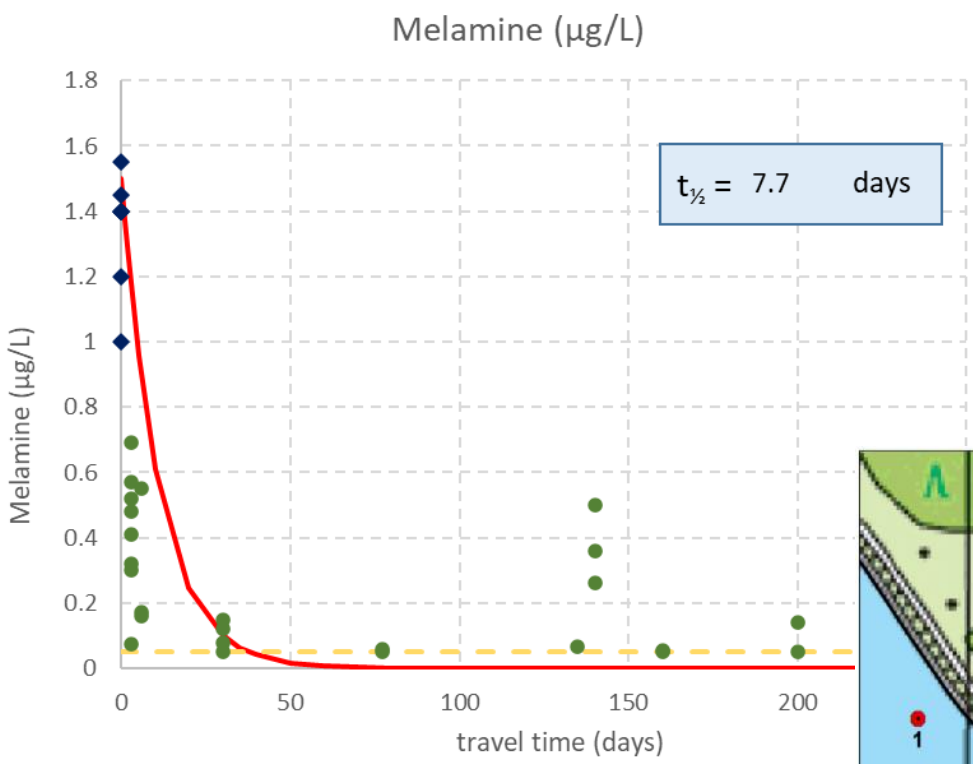
Verwijdering organisch microverontreinigingen tijdens bodempassage

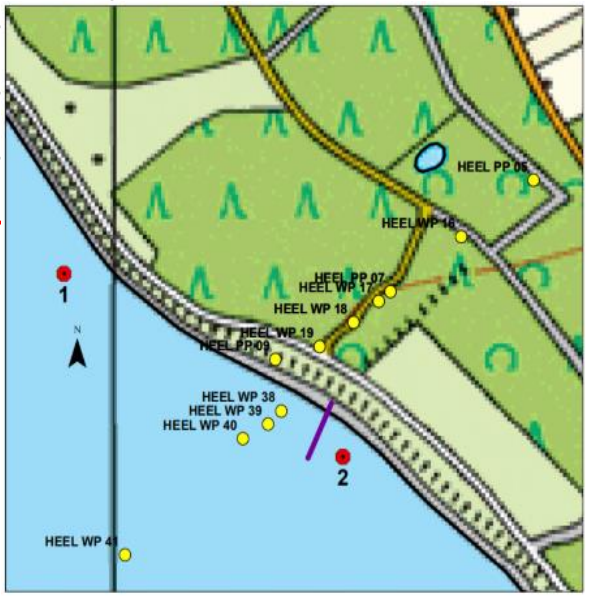
Concentratie in spaarbekken en pompput



OMV verwijdering langs stroombaan

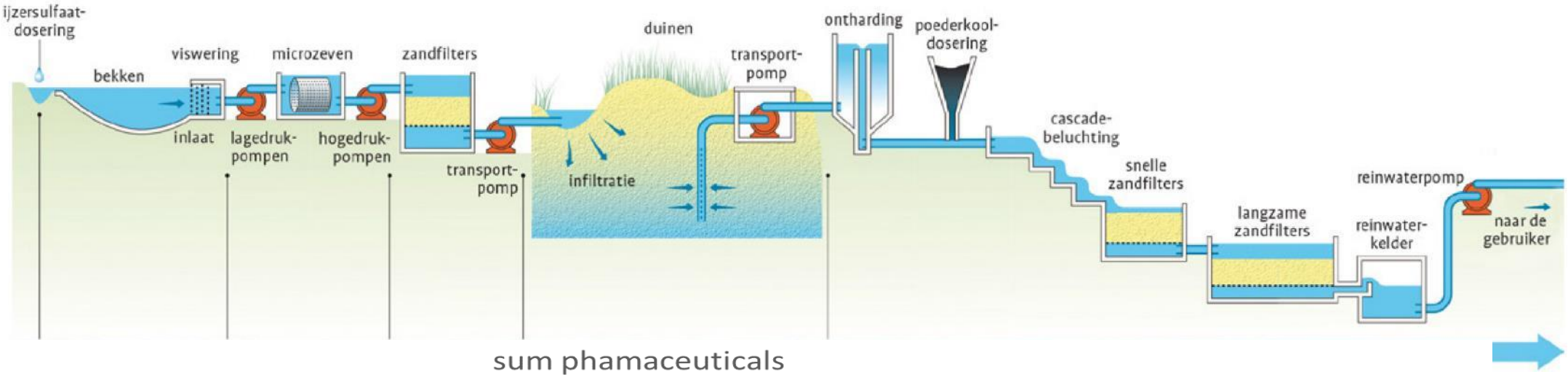




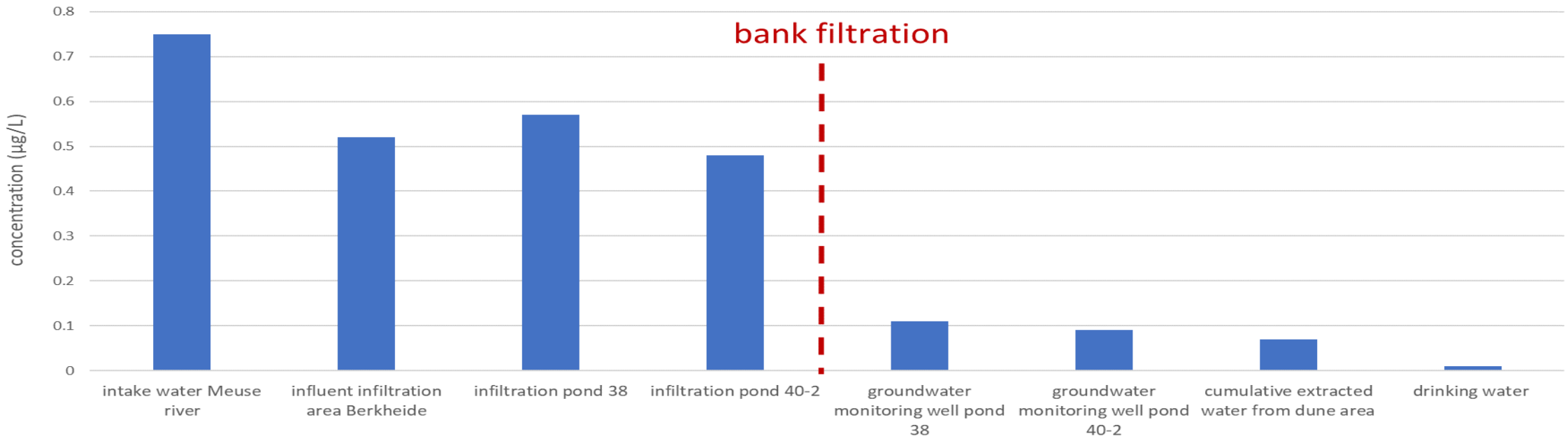


Drinking water production from river water

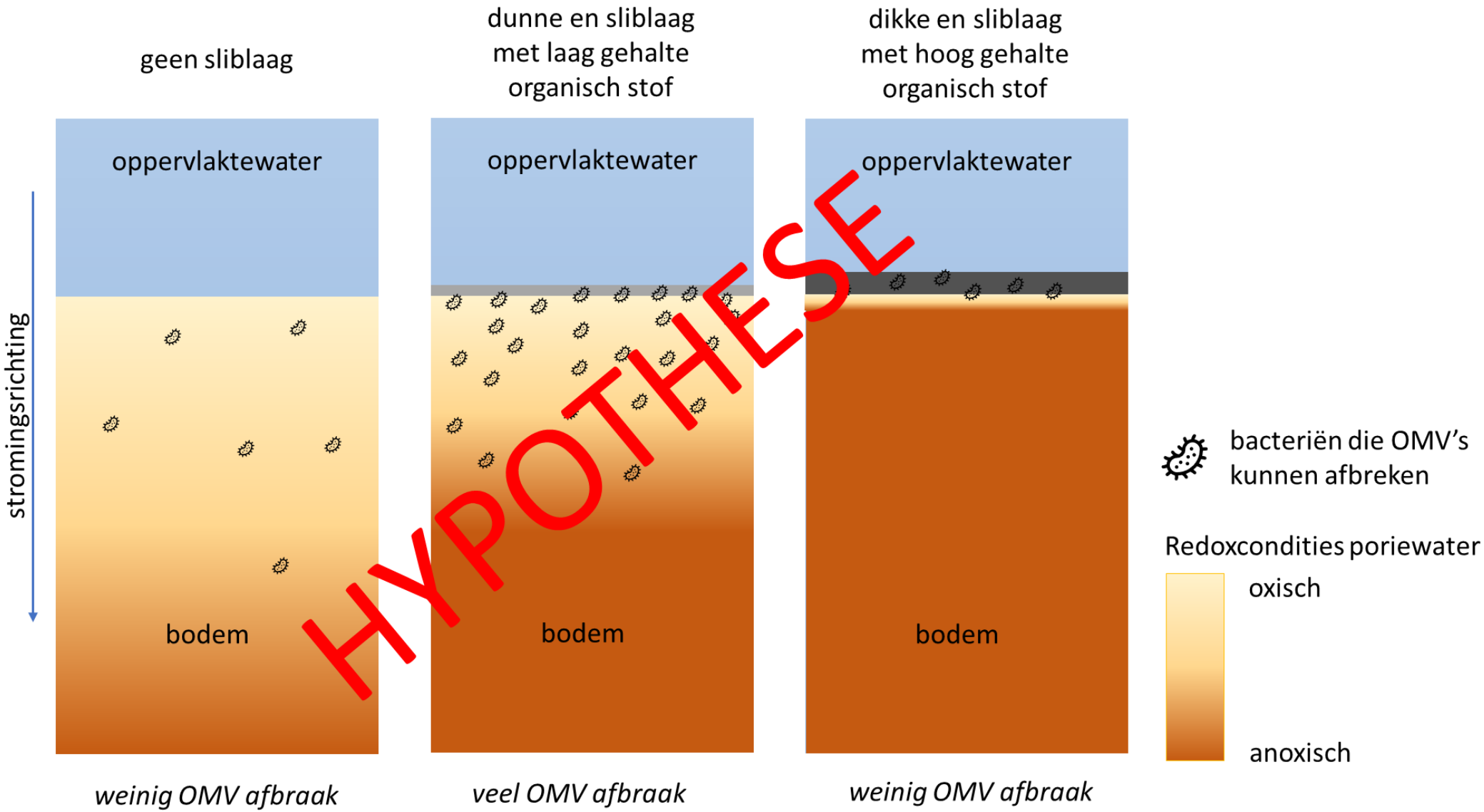
river → pre-treatment → basis artificial recharge → post-treatment → drinking water



sum phamaceuticals



Effect sliblaag

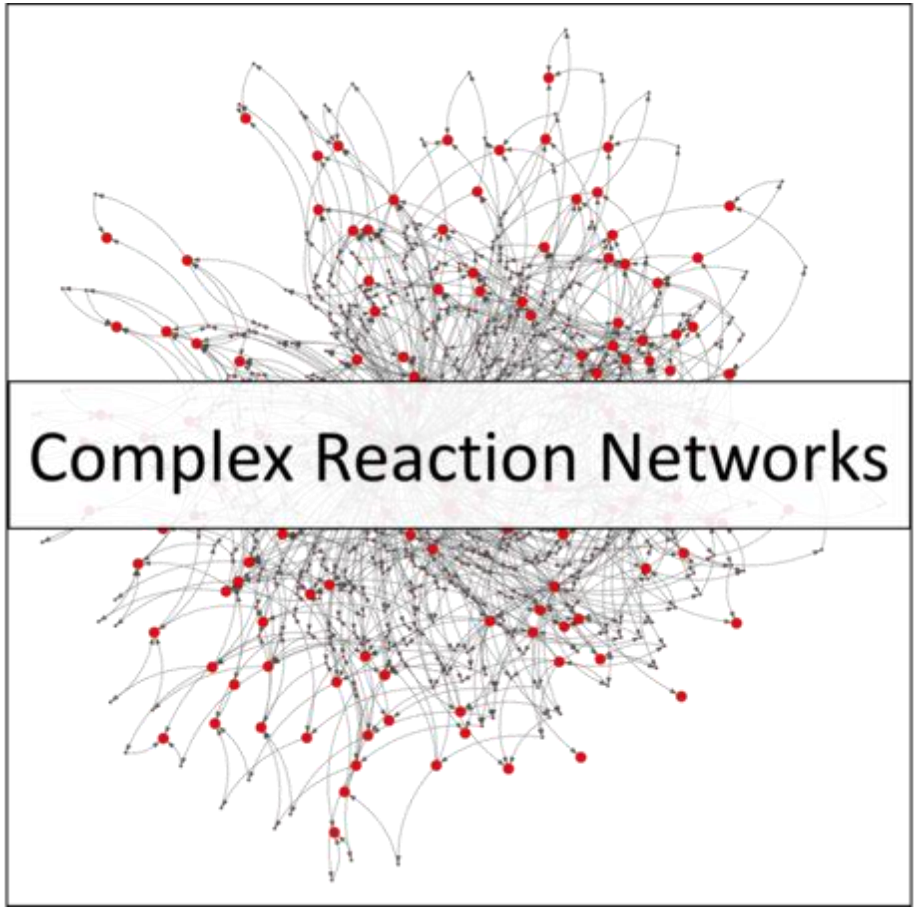


Stofeigenschappen en inschatten hiervan: Complexiteit!

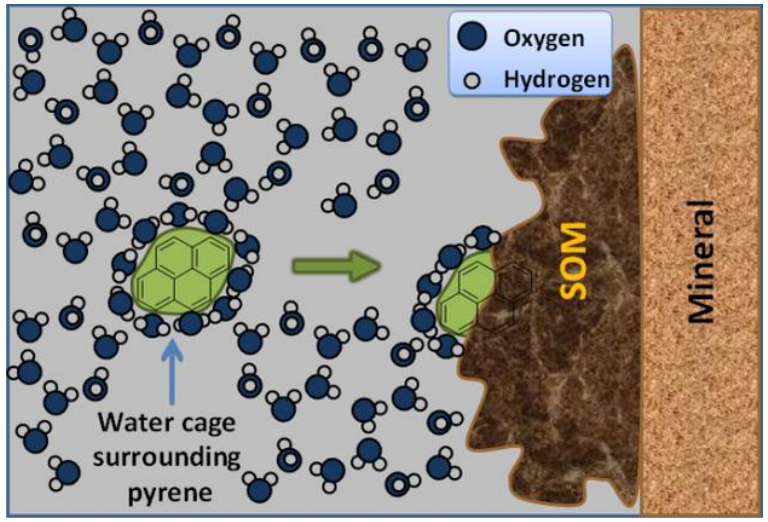
- Heel veel stoffen!!
- Halfwaardetijden?
- Zijn mobiele stoffen wel mobiel? (polaire geïoniseerde omv's)

indien we de processen begrijpen kunnen we die potentieel gebruiken bij ontwerp infiltratie

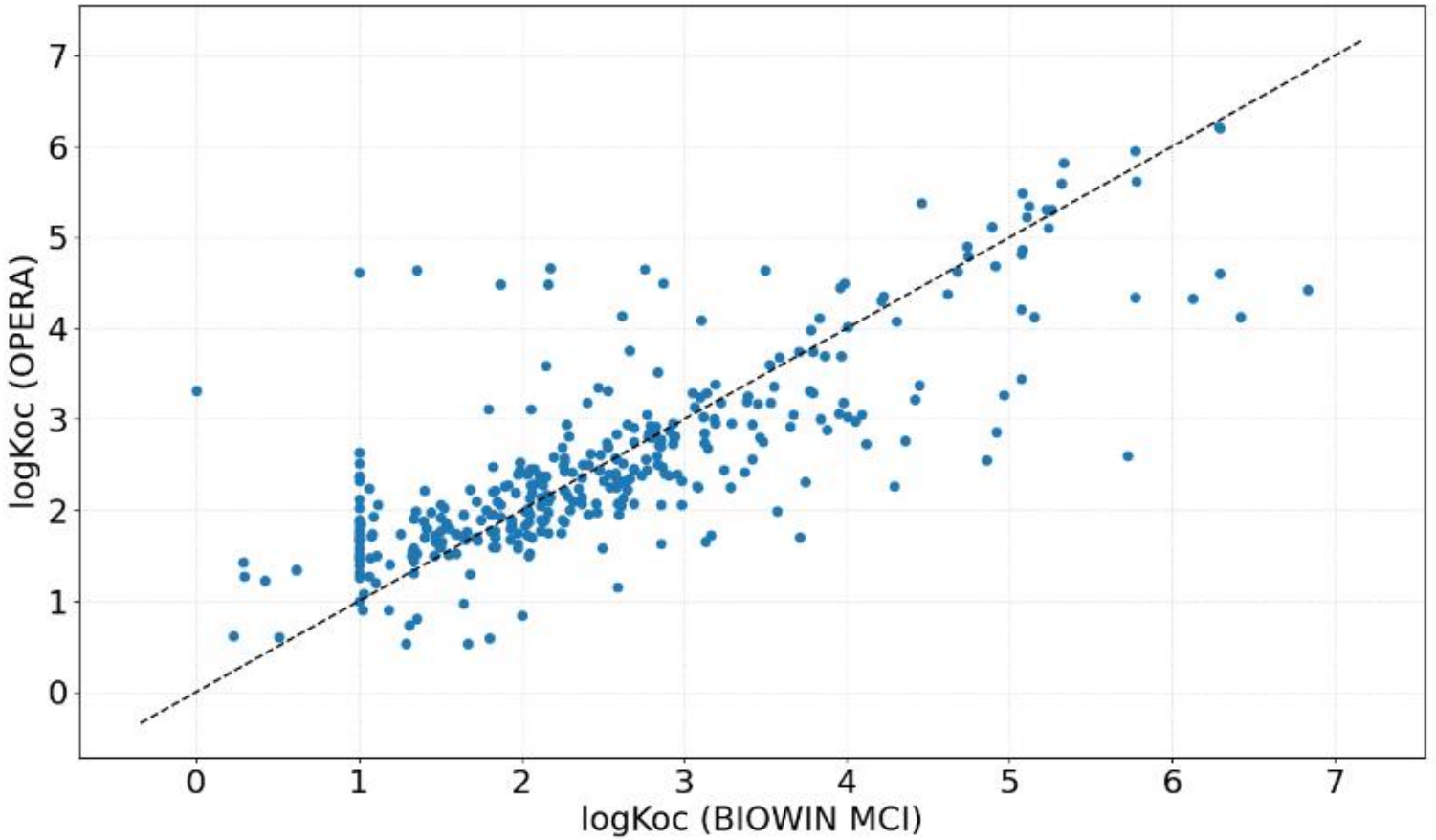
- Bijvoorbeeld: zeolieten, ijzerzand, ect
- Regeneratie zuiveringsmaterialen



Mobiliteit
Organisch koolstof partitiecoëfficiënt: K_{OC}



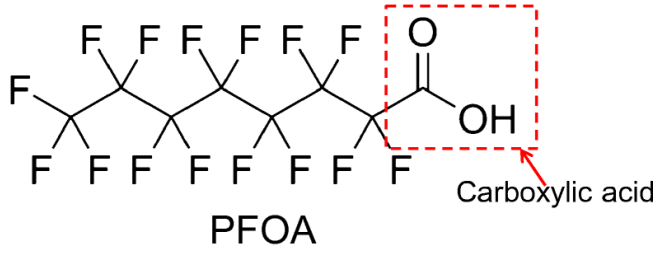
$Kd = K_{OC} \times f_{OC}$



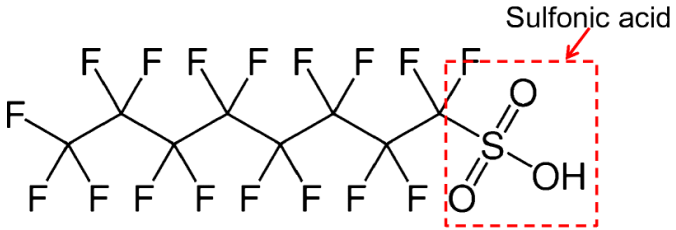
Zijn mobiele stoffen wel mobiel?

Veel organische microverontreinigingen zijn polair of geladen

Als anion

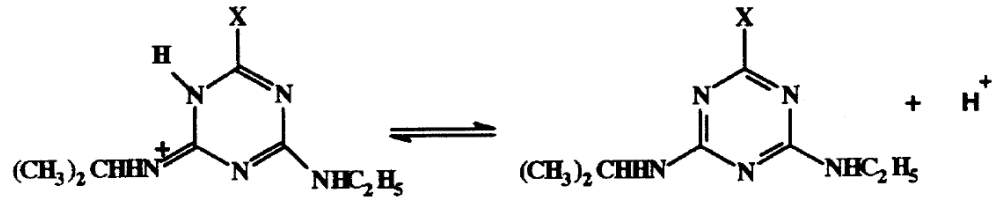


PFOA



PFOS

Als kation



If X = Cl, pKa = ~2
If X = OH, pKa = ~5

Charge of reactive soil surfaces

In the environment we find reactive solids with a large, charged surface area and the ability to bind solutes. A few main ones:

(Hydro)oxides, iron-oxides

Clay

Organic matter

Adsorptie van geladen OMVs aan bodemdeeltjes wordt in huidige praktijk onderschat: nieuw model nodig

Persistentie/afbraak
Halfwaardetijd

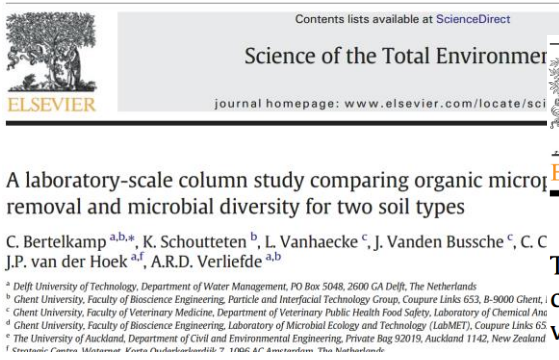
- Veldwaarnemingen
- Literatuurwaardes

Arp & Hale (2022): voor slechts 2.2% van de 14.203 chemische stoffen in REACH zijn (laboratorium) halfwaardetijden bekend uit gestandaardiseerde proeven

Gebruik van op QSAR gebaseerde modellen voor biodegradatie zoals EpiSuite en OPERA

Database with compound properties

- OMP database with 355 compounds from literature - in different conditions, resulting in 1239 observations
- Field data from monitoring transects in the Netherlands
- Chemical properties: $\log K_{OW}$, $\log K_{oc}$, pKa and T½.
- Standard chemical properties: type of compound, the molar mass and solubility
- Meta-information: type of experiment, redox condition, type of sediment used, concentration range of the OMP, ….



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Science of the Total Environmer

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A laboratory-scale column study comparing organic micropl
removal and microbial diversity for two soil types

C. Bertelkamp [a,b,*], K. Schoutteten [b], L. Vanhaecke [c], J. Vanden Bussche [c], C. C
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[d] Ghent University, Faculty of Bioscience Engineering, Laboratory of Microbial Ecology and Technology (LabMET), Coupure Links 65
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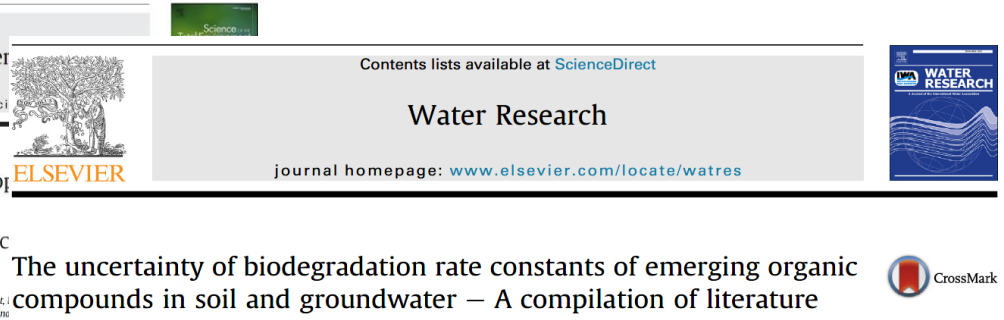

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Water Research

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The uncertainty of biodegradation rate constants of emerging organic compounds in soil and groundwater – A compilation of literature values for 82 substances

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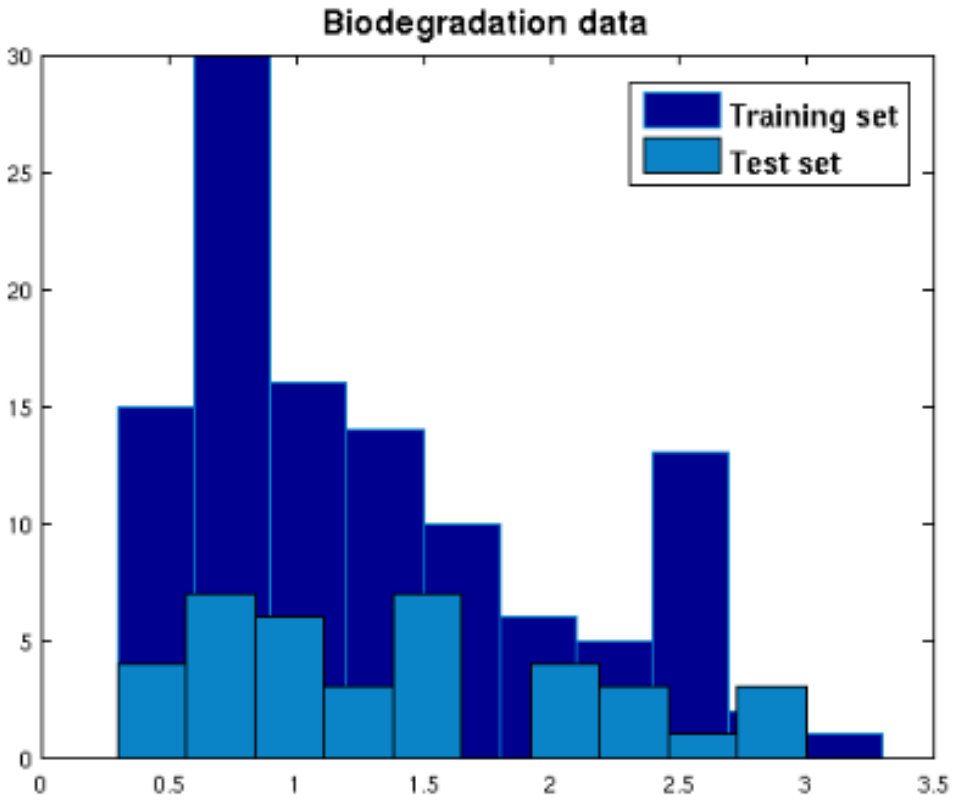
ABSTRACT

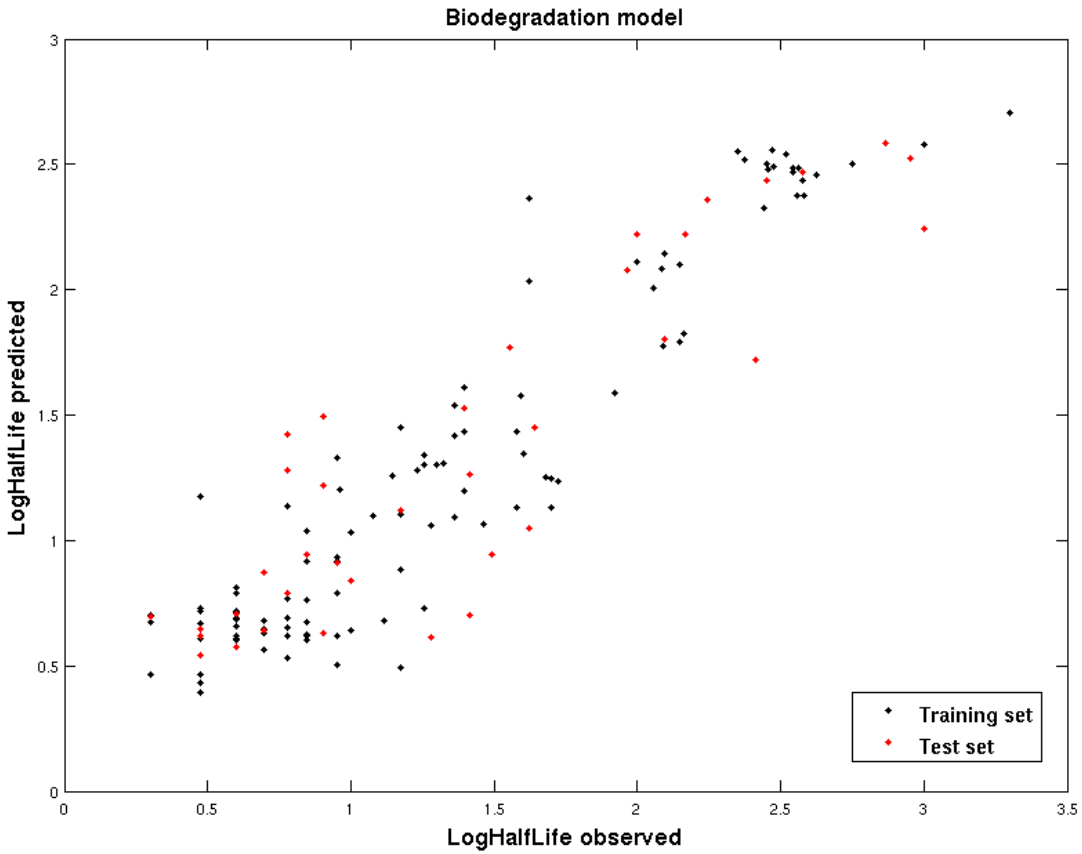
The present study reports on biodegradation rate constants of emerging organic compounds (EOCs) in soil and groundwater available in the literature. The major aim of this compilation was to provide an assessment of the uncertainty of hydrological models with respect to the fate of EOCs. The literature search identified a total number of 82 EOCs for which 1st-order rate constants could be derived. It was found that for the majority of compounds degradation rate constants vary over more than three orders of magnitude. Correlation to factors that are well known to affect the degradation rate, such as temperature or redox condition was weak. No correlation at all was found with results from available quantitative structure-activity relationship models. This suggests that many unknown site specific or experimentally

Own KWR database (Pieter Stuyfzand)

Substance	CAS-No.	Log K_{ow} (KOWWIN v1.67 estimate)	Log K_{ow} (experimental database)	Log K_{oc} (PCKOCWIN v1.66)	pK_a (dissociation constant)	Minimum rate constant (1/d)	Maximum rate constant (1/d)	Rate co	Concentration range OMP (µg/L)	Type of experiment
Carbendazim	10605-21-7	1.55	1.52	2.245	4,29; 4,2	persistent	persistent		0 - 5	Field investigati
Diglyme	111-96-6	-0.48	-0.36	0.000	-	persistent	persistent		5.00E-01	Column experim
Diglyme	111-96-6	-0.48	-0.36	0.000	-	persistent	persistent		0 - 5	Field investigati
Diglyme	111-96-6	-0.48	-0.36	0.000	-	persistent	persistent		0 - 5	Field investigati
Tris(2-chloroethyl)phosphate (TCEP)	115-96-8	1.63	1.44	2.478	-	persistent	persistent		0,02 - 1,5	Column experim
Tris(2-chloroethyl)phosphate (TCEP)	115-96-8	1.63	1.44	2.478	-	persistent	persistent		4.00E-01	Field investigati
Amidotrizoic / Diatrizoic acid	117-96-4	-	-	-	1,13; 7,95	persistent	persistent		0 - 5	Field investigati
1,4-Dioxane	123-91-1	-0.32	-0.27	0.000	-	persistent	persistent		7.90E-01	Field investigati
1,4-Dioxane	123-91-1	-0.32	-0.27	0.000	-	persistent	persistent		0 - 5	Field investigati
1,4-Dioxane	123-91-1	-0.32	-0.27	0	-	persistent	persistent	persistent	0 - 1,8	Field investigati
1,4-Dioxane	123-91-1	-0.32	-0.27	0.000	-	persistent	persistent		0 - 5	Field investigati
Primidone	125-33-7	0.73	0.91	2.855	12,3	persistent	persistent		1.50E-01	Field investigati
Primidone	125-33-7	0.73	0.91	2.855	12,3	persistent	persistent		-	Tank experiment (no s
Primidone	125-33-7	0.73	0.91	2.855	12,3	persistent	persistent		4.60E-01	Tank experiment (no s
Primidone	125-33-7	0.73	0.91	2.855	12,3	persistent	persistent		4.00E+01	Batch experime
Primidone	125-33-7	0.73	0.91	2.855	12,3	persistent	persistent		0.04-0.49	Field investigati
Primidone	125-33-7	0.73	0.91	2.855	12,3	persistent	persistent		0 - 0,64	Field investigati
Primidone	125-33-7	0.73	0.91	2.855	12,3	persistent	persistent		4.00E+01	Batch experime
N,N-Diethyl-meta-toluamide (DEET)	134-62-3	2.26	2.18	2.730	-	persistent	persistent		3.20E-01	Field investigati
Tris(1-chloro-2-propyl)phosphate (TCPP)	13674-84-5	2.89	2.59	3.107	-	persistent	persistent		0,02 - 1,5	Column experim
Diclofenac	15307-86-5	4.02	4.51	2.921	4,15	persistent	persistent		0,10 - 0,96	Column experim
Diclofenac	15307-86-5	4.02	4.51	2.921	4,15	persistent	persistent		1.00E+01	Column experim
Lincomycin	154-21-2	0.29	0.56	1.768	7,6	persistent	persistent		2.00E-01	Column experim
4-Formylaminoantipyrin (FAA)	1672-58-8	0.50	-	1.804	-	persistent	persistent		0,10 - 0,96	Column experim
Clindamycin	18323-44-9	2.01	2.16	1.768	-	persistent	persistent		1.20E+00	Tank experiment (no s
Atrazine	1912-24-9	2.82	2.61	2.362	1,60	persistent	persistent		20 - 174	Batch experime
Atrazine	1912-24-9	2.82	2.61	2.362	1,60	persistent	persistent		1.00E+01	Closed-loop fixed bed
Atrazine	1912-24-9	2.82	2.61	2.362	1,60	persistent	persistent		2.00E-01	Column experim
2,6-dichlorobenzamide (BAM)	2008-58-4	0.90	0.77	2.149	-	persistent	persistent		2.50E+01	Column experim
Bentazon	25057-89-0	1.67	2.34	1.574	3,3	persistent	persistent		2.50E+01	Column experim

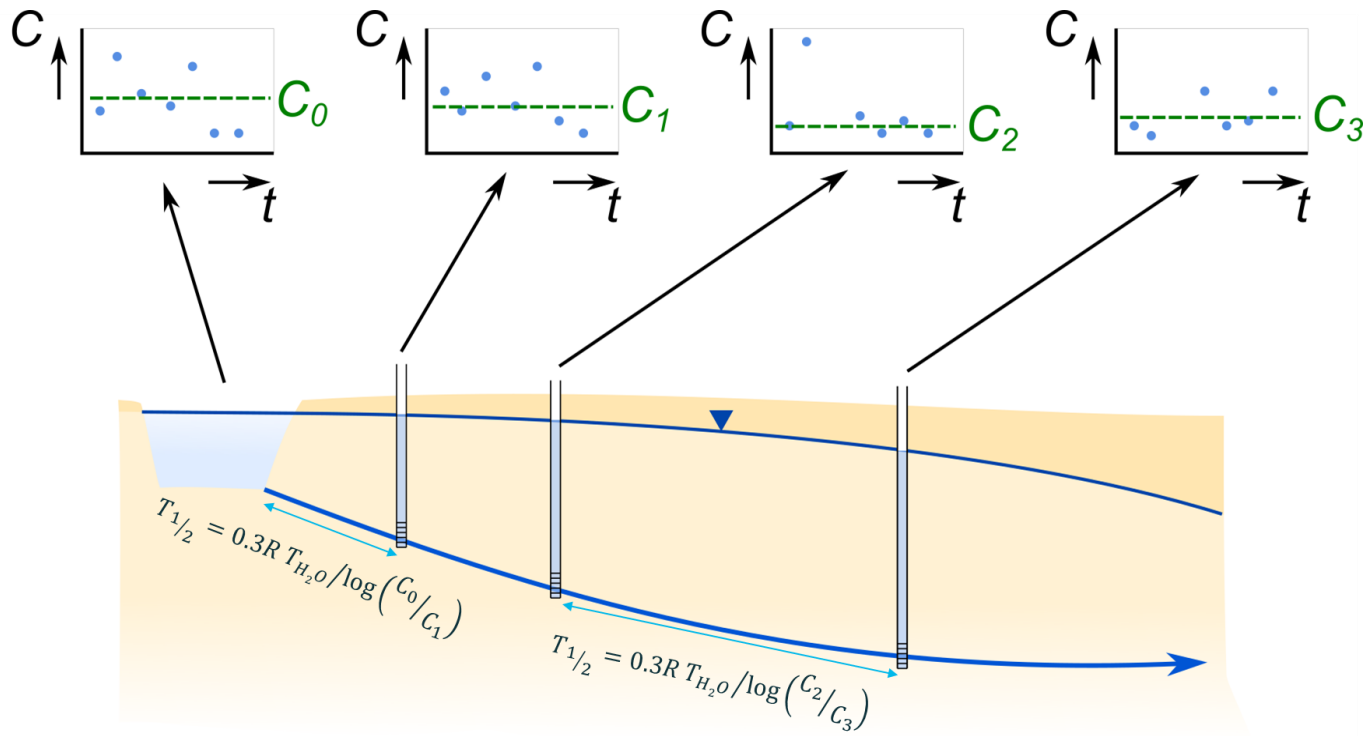
OPERA Biodegradatie model



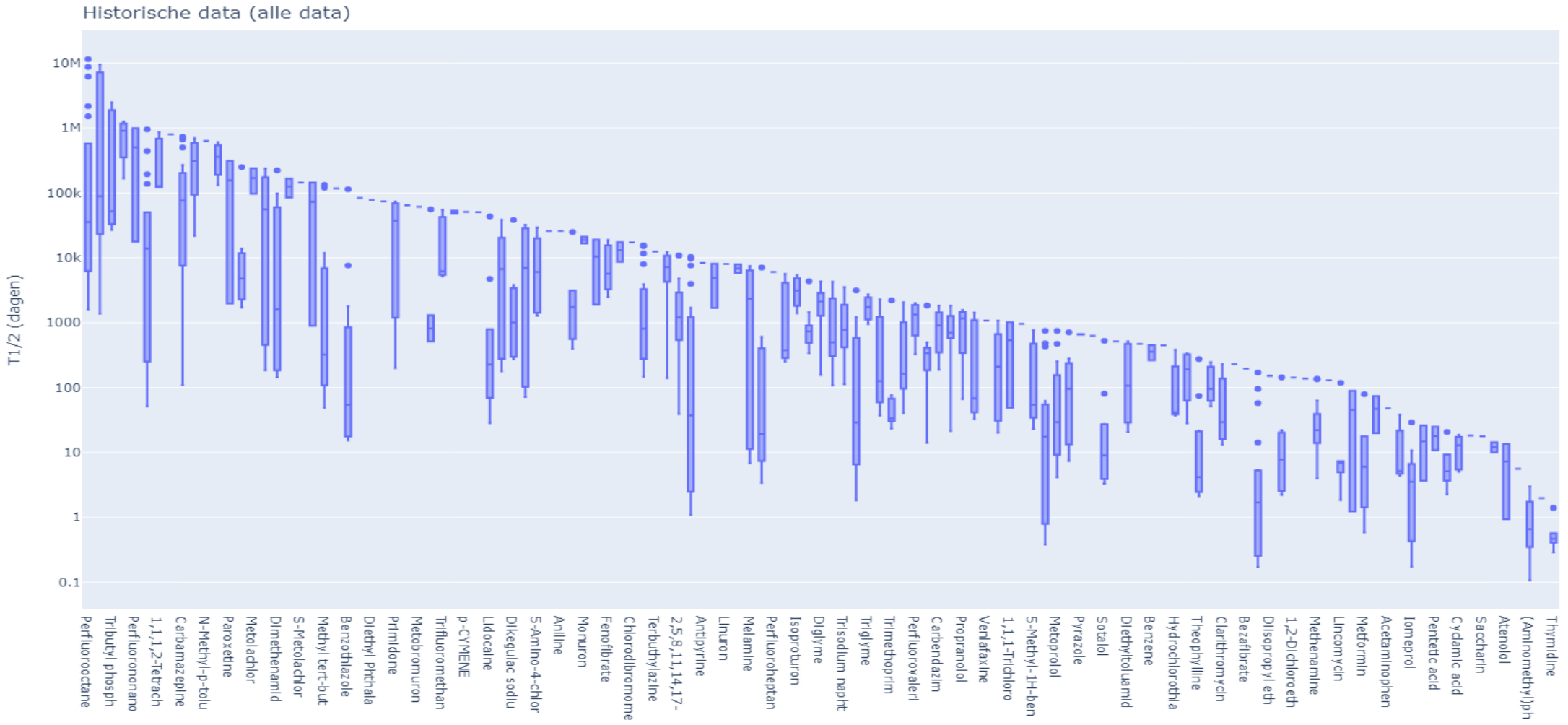


Field data on degradation organic micro-pollutants during bank filtration

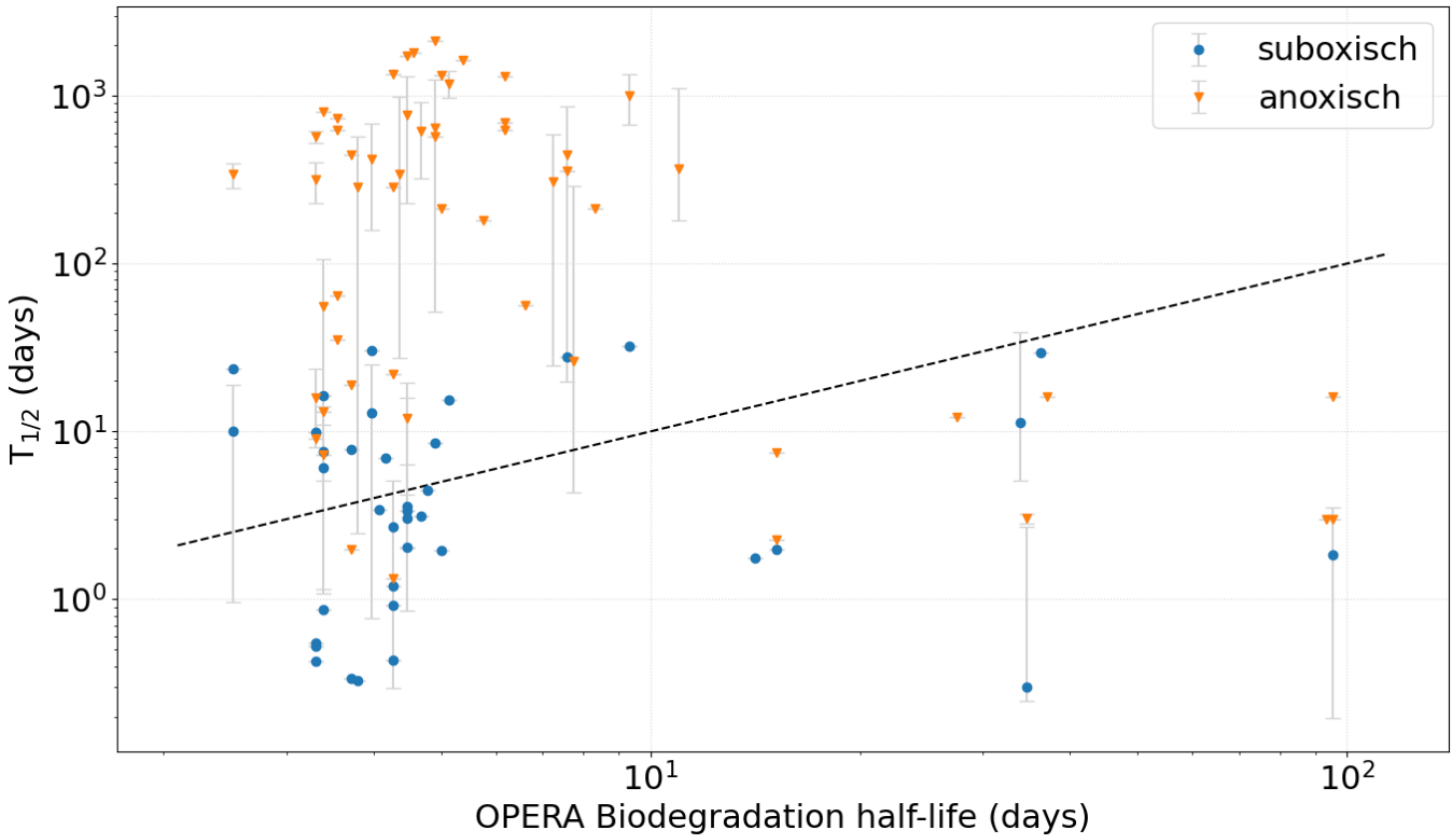
- 8 Monitoring transects across the Netherlands with known travel times
- Riverbank filtration and basin infiltration sites
- Historic data drinking water utilities
- Additional monitoring campaign
- 183 compounds with one or more T½ values



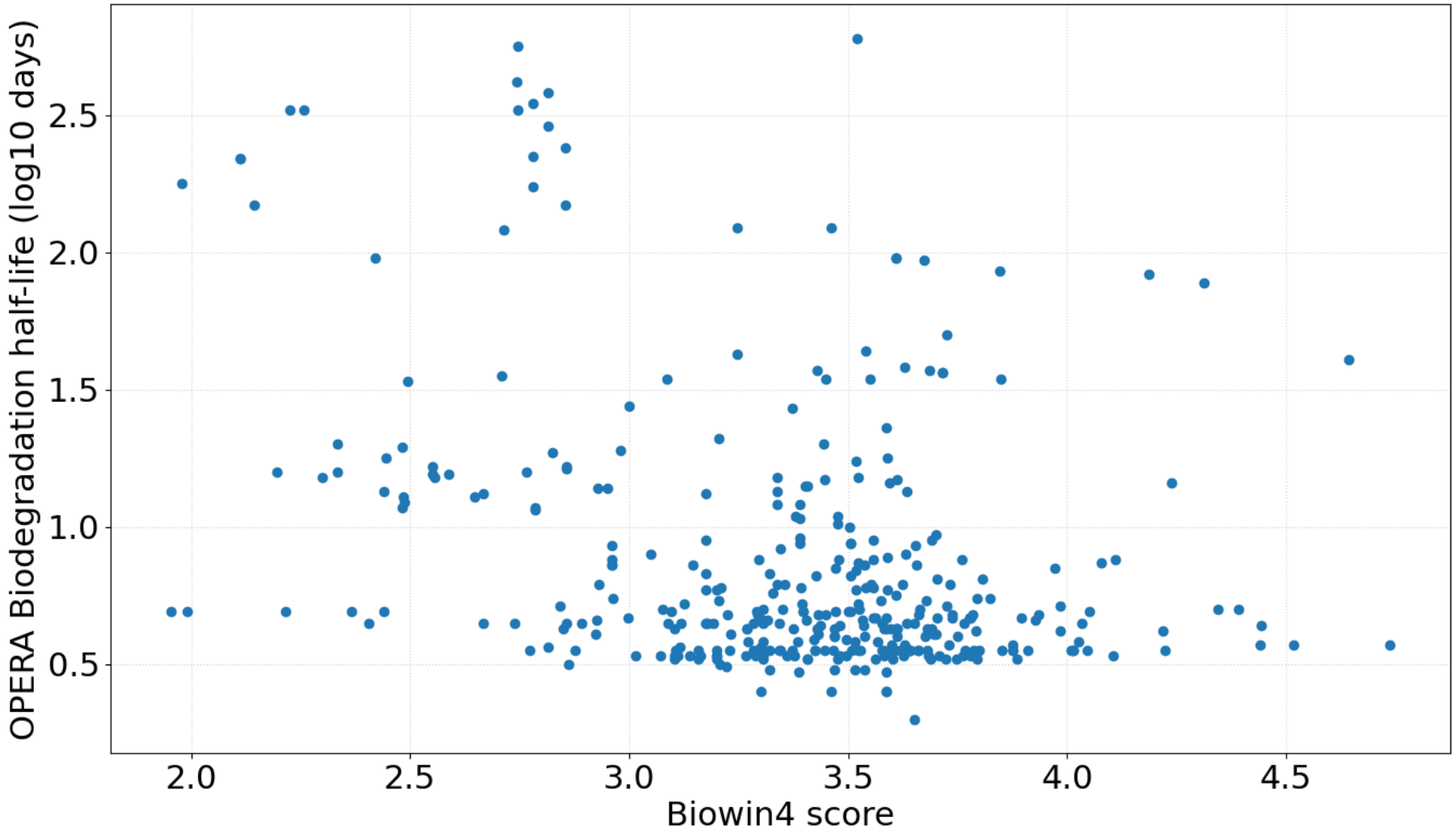
Halfwaardetijden uit data meetraaien



Vergelijking gemeten halfwaardetijden in grondwatersystemen met database



Existing databases do not agree as well…

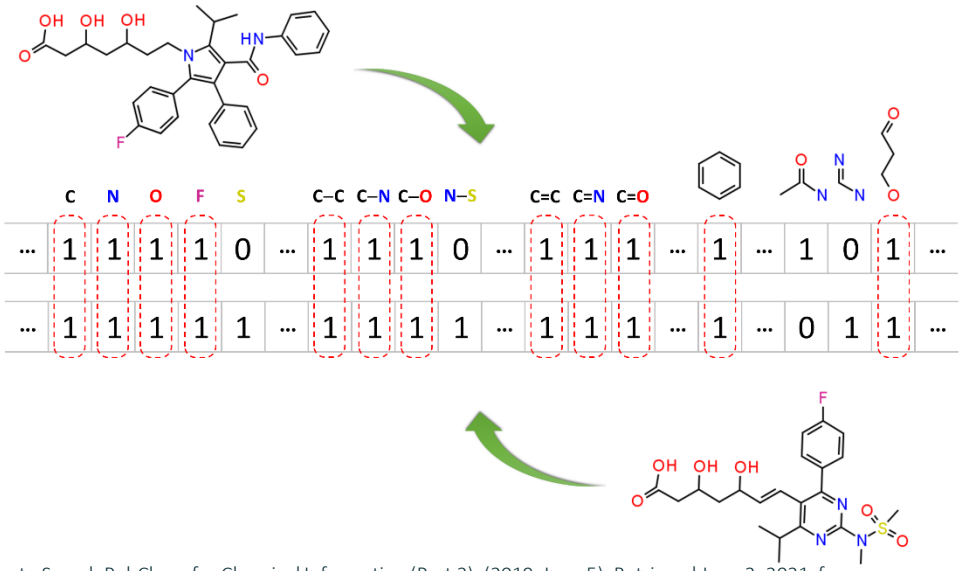


Clustering of SVHCs

1. Start with compounds lists (SVHCs list, pSVHCs list, PMT list, ..) >1000 compounds
2. Cluster compounds in representative clusters
   - Using chemical descriptors
   - Statistical model
   - Evaluate number of clusters
3. Choose relevant compounds from clusters
4. Make list for target analyses:
   - ~30-40 compounds
   - Preferably within one analysis method (to reduce costs)

Clustering – compound properties

- From pubchem → isomeric smiles
- Fingerprints:
  - CACTVS fingerprint
  - MACCS fingerprint

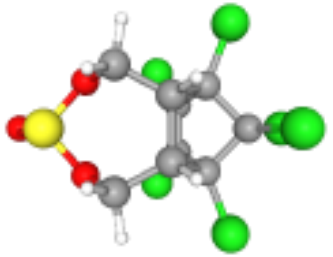


How to Search PubChem for Chemical Information (Part 2). (2019, June 5). Retrieved June 3, 2021, from https://chem.libretexts.org/@go/page/95659

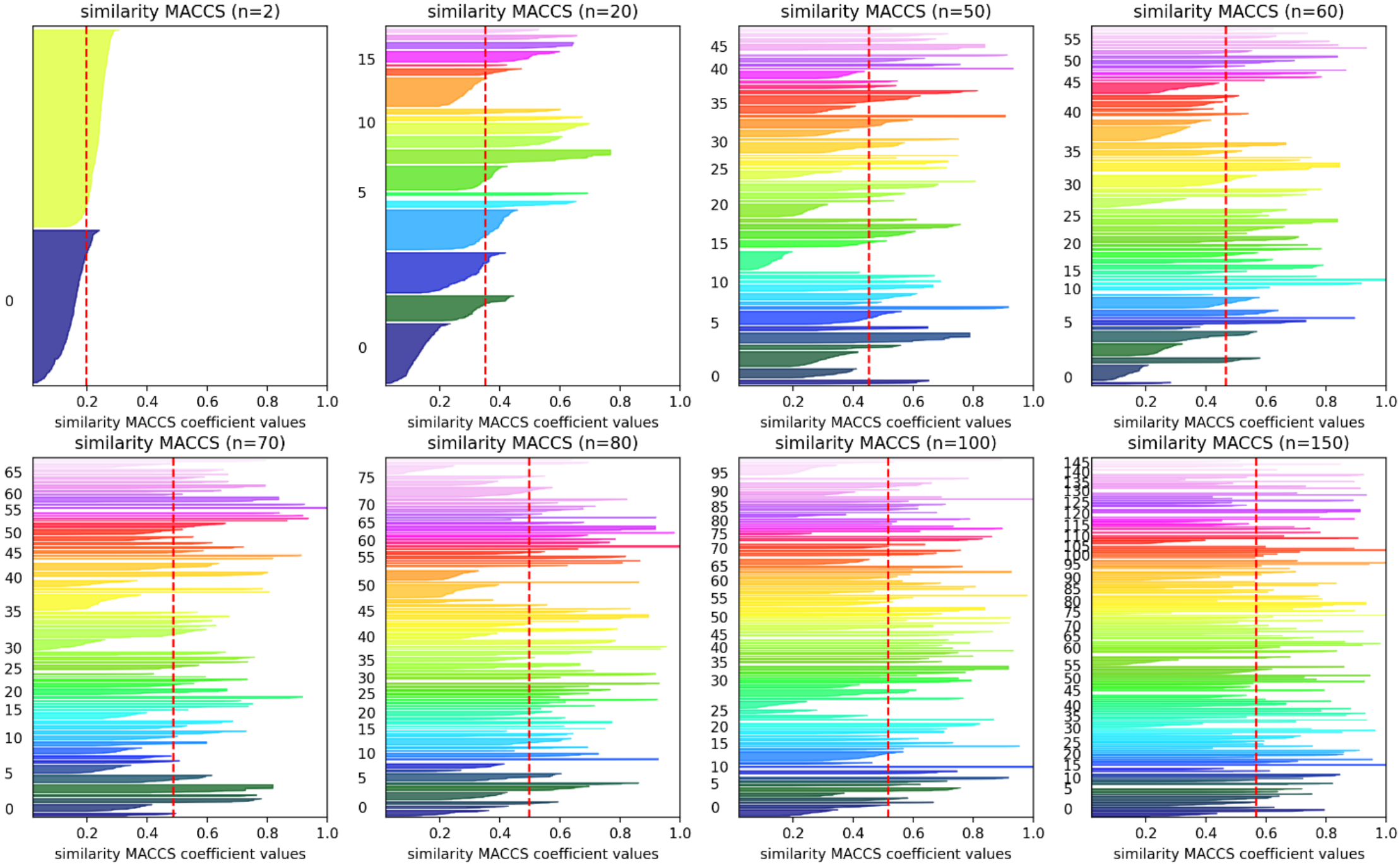
- Descriptors pubchem:logp, charge, tpsa, rotatable_bond, molecular_weight, heavy_atom, h_bond_acceptor, h_bond_donor

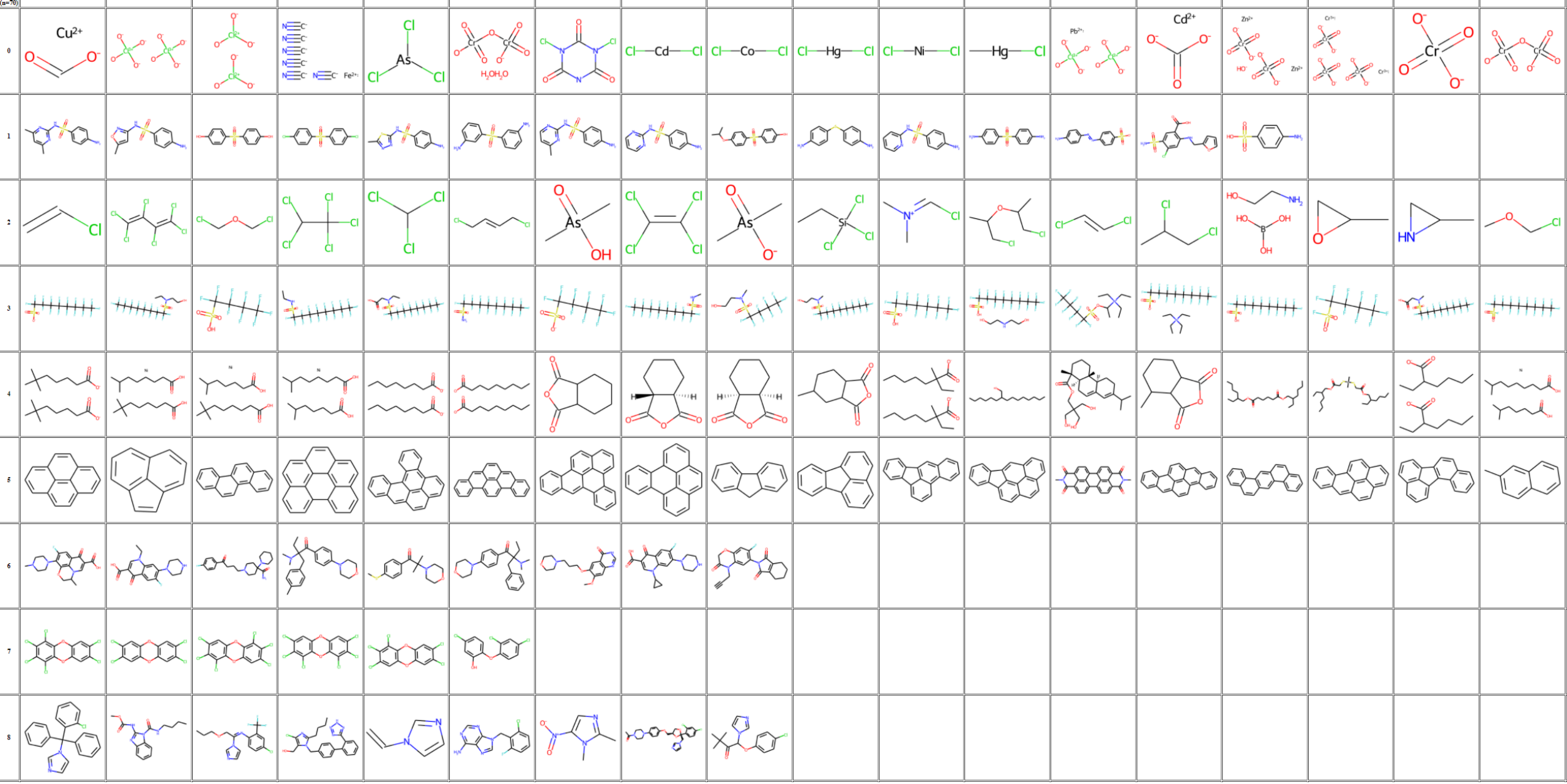
Example

115-29-7 (endosulfan)



C1C2C(COS(=O)O1)C3(C(=C(C2(C3(Cl)Cl)Cl)Cl)Cl)Cl


similarity MACCS (n=2)
similarity MACCS (n=20)
similarity MACCS (n=50)
similarity MACCS (n=60)
similarity MACCS (n=70)
similarity MACCS (n=80)
similarity MACCS (n=100)
similarity MACCS (n=150)
similarity MACCS coefficient values


cluster (n=70)
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17

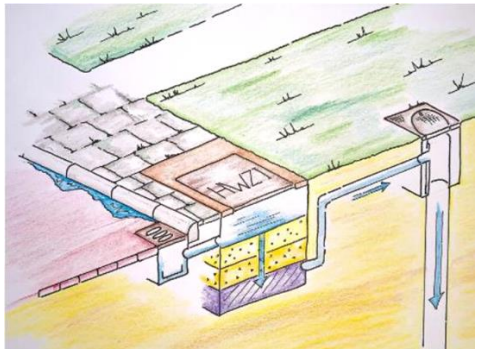
Select compounds per cluster (bold in analysis method)

cluster	Stofnaam (hoogst geprioriteerd per cluster)
0	trichloro(fluoro)methane
1	**Furosemide**
2	Chloroform
3	**1,1,2,2,3,3,4,4,4-nonafluorobutane-1-sulfonic acid (PFBS)**
4	**Cycloheximide**
5	Phenanthrene
6	Flumioxazin
8	**Climbazole**
9	**2-hydroxybenzothiazool**
10	**2-methylbenzothiazool**
11	**Nicosulfuron**
12	**Pentoxyfylline**
13	1,2-dibroomethaan
14	**Benzotriazole**
16	**2,4-dinitrofenol**
17	**Terbutylazine**
18	Endrin
19	Iopamidol
20	**Metoprolol**
21	**2,4,5-trimethylaniline**
24	**Carbendazim**
25	**triethyl phosphate**
26	parathion-methyl
28	Prothioconazool
29	**Thiacloprid**
30	**Phoxim**

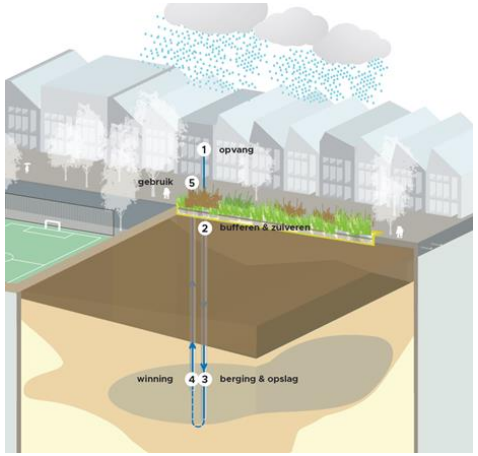
cluster	Stofnaam (hoogst geprioriteerd per cluster)
34	**2-(benzotriazol-2-yl)-4-methylphenol (Drometrizole)**
35	pentadecafluorooctanoic acid
37	**Methamphetamine**
39	N-[bis[(butan-2-ylideneamino)oxy]-methylsilyl]oxybutan-2-imine
40	**Methadon**
41	**CGA 354743 (Metolachlor esa)**
42	**Warfarin**
43	3-(oxiran-2-ylmethoxy)-N,N-bis(oxiran-2-ylmethyl)aniline
44	**benzoylecgonine (metaboliet cocaine)**
46	**Tetraglyme**
47	dimethoxy(dimethyl)silane
48	Tetrahydrofuran
51	**Metobromuron**
52	1,2,4-trichlorobenzene
55	**Bromacil**
56	**Saccharine**
57	**MDMA**
59	[bis[6-[bis(phosphonomethyl)amino]hexyl]amino]methylphosphonic acid
60	**Metazachloor**
61	Salicylzuur
62	beta-endosulfan
64	**Oxazepam**
65	Hexanedihydrazide
67	**4-chloor-2-methylfenoxyazijnzuur (MCPA)**
69	**Oxybenzone**

TKI-project Straatwaterfiltratie voor infiltratie (2024-2027)

Doel: Ontwikkelen praktisch haalbare en betrouwbare methode voor beschermen van de grondwaterkwaliteit bij infiltratie van afstromend hemelwater



- Beter zicht op voor grondwaterkwaliteit relevante verontreinigingen in afstromend hemelwater
- Beter zicht op transportgedrag van deze stoffen in bodem en grondwater
- Beter zicht op (toekomstige) wet- en regelgeving
- Kansrijke filtermedia (ook vanuit hydraulisch oogpunt)
- Geschikte ontwerpen voor verschillende vormen van infiltratie



TKI-project Straatwaterfiltratie voor infiltratie (2024-2027)

Consortium: