Supplemental Materials for the paper: Pesticide exposure in small streams in Germany

Eduard Szöcs, Marvin Brinke, Bilgin Karaoglan, Ralf B. Schäfer

Contents

1	Data Cleaning	5
2	Overview on compiled data	7
3	Thresholds for agricultural land use and catchment size	22
4	Effect of precipitation and season on RQ	23
5	Pesticides in small streams	28

List of Figures

S1	Overview on data cleaning steps	6
S2	Number of sampling occasions per year and month	8
S3	Complete Linkage Cluster Dendrogram of Jaccard Similarity of analysed	
	compound spectra between federal states	20
S4	Average silhouette width for different cluster sizes	21
S5	Raw data used for the model in equation 2 and Figure 3 of the main article.	22
S6	Distribution of precipitation at sampling occasions	26
S7	Graphical representation of coefficients from table S4	27
S8	Cumulative distribution of the number sites exceeding RAC	30
S9	Proportion of samples with detects in small streams	31
S10	Distribution of the number of quantified compounds in the samples	32

List of Tables

S1	Overview on chemical samples	7
S2	Overview on pesticides in the database	8
S3	23 pesticides for which we modelled the relationship with precipitation and seasonality	23
S4	Coefficients and CI from per compound models	
S5	Overview on RAC exceedances of the 76 compounds with more than 1000	20
	measurements.	-28

1 Data Cleaning

Before combining into a common database, more than 30 datasets have been cleaned and homogenized separately. Cleaning steps comprised the following steps (Figure S1 gives a graphical overview):

- 1. Structure: Datasets have been adjusted to the database structure.
- 2. Coordinates: Coordinates have been transformed to a common Coordinate Reference System (DHDN / 3-Grad Gauss-Krüger Zone 3 (EPSG:31467)) and duplicates merged.
- 3. Chemicals: Chemical names and identifiers have been unified using the webchem package (https://github.com/ropensci/webchem).
- 4. Identifiers: Unique identifiers have been assigned.
- 5. Units: All concentrations have been converted to $\mu g/L$. Values below limit of quantification were set to zero (and can be used to identity non-detects).
- 6. Other meta-data: meta-data has been standardised.
- 7. Temporal resolution: The temporal resolution of the database is 1 day. Samplings below this resolution have been aggregated by the maximum daily value.
- 8. Validity Checks: Simple rules for validity checks have been implemented.



Figure S1: Overview on data cleaning steps. After cleaning, data has been stored in a relational spatial PostgreSQL database.

2 Overview on compiled data

Table S1: Overview on chemical samples. Only data from running waters and grab sampling is shown. ^a: Abbreviations according to ISO 3166-2:DE. ^b: Including metabolites

name	abbrv.ª	Begin	End	No. sites	No.samples	No. pesticides ^b
Baden-	BW	2005-03-10	2014-10-02	7	172	98
Württemberg						
Bavaria	BY	2006 - 04 - 19	2013 - 12 - 17	13	218	155
Hesse	$_{ m HE}$	2007 - 01 - 15	2014-12-18	65	2411	144
Mecklenburg-	MV	2005-03-08	2014 - 12 - 17	130	1503	227
Western						
Pomerania						
Lower Saxony	NI	2014-03-24	2014 - 10 - 13	1	7	226
North Rhine-	NW	2005-01-18	2015 - 01 - 22	1139	8536	198
We stphalia						
Rhineland-	RP	2008-01-02	2013-12-18	7	341	236
Palatinate						
Schleswig-	SH	2005-04-26	2014 - 11 - 26	269	1380	180
$\operatorname{Holstein}$						
Saarland	SL	2005 - 01 - 03	2013 - 11 - 25	2	104	57
Saxony	SN	2005 - 01 - 02	2013-12-18	606	9141	173
Saxony-Anhalt	ST	2005 - 01 - 24	2015 - 03 - 19	30	416	88
Thuringia	TH	2005-06-16	2014-12-08	32	514	63
	Total	2005-01-02	2015-03-19	2301	24743	478

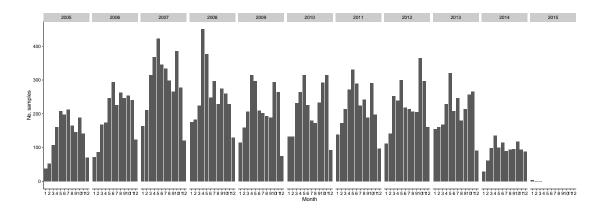


Figure S2: Number of sampling occasions per year and month.

Table S2: Overview on pesticides (and metabolites) in the database. ^a Authorized in Germany (Source: German Federal Office of Consumer Protection and Food Safety (BVL) as at March 2015). ^b Authorized in the European union (Source: EU Pesticides database as at March 2015). ^c Regulatory Acceptable Concentration $[\mu g/L]$ (Source: German Environment Agency (UBA) as at November 2015).

	Name	CAS	Group	${ m Auth.} \ { m GER^a}$	${ m Auth.} \ { m EU^b}$	RAC ^c
1	(E)7-(Z)9-	55774-32-8	other	X	X	
	Dodecadienylacetat					
2	(Z)-9-Dodecenylacetat	16974-11-1	other	X	X	
3	1,3-cis-Dichlorpropen	10061 - 01 - 5	other			
4	1,3-trans-Dichlorpropen	10061-02-6	other			
5	1-(3,4-	2327-02-8	${ m metabolite}$			
	Dichlorphenyl)urea					
6	1-(4-	56046 - 17 - 4	${ m metabolite}$			
	${\rm Isopropylphenyl) urea}$					
7	1-Decanol	112 - 30 - 1	other	X	X	
8	1-Methylcyclopropen	3100 - 04 - 7	other	X	X	
9	2,4,5-T	93 - 76 - 5	herbicide			
10	2,4,6-Trichlorphenol	88-06-2	${ m metabolite}$			
11	2,4-D	94 - 75 - 7	herbicide	X	X	1.10000
12	2,4-DB	94-82-6	herbicide		X	
13	2,4-Dichlorphenol	120 - 83 - 2	${ m metabolite}$			
14	2,6-Dichlorobenzamid	2008-58-4	${ m metabolite}$			
15	2-	19988-24-0	${ m metabolite}$			
	Hydroxydesethylatrazin					
16	3-Hydroxy Carbofuran	16655 - 82 - 6	${ m metabolite}$			
17	4,6-Dinitro-o-Cresol	534 - 52 - 1	in sectic ide			

18	4-tert. Cyclobutylhex-	98-53-3	${ m metabolite}$			
1.0	anon	1000 51 0	. 1 11.			
19	AMPA	1066-51-9	metabolite			0.00000
20	Acequinocyl	57960-19-7	insecticide	X	X	9.00000
21	Acetamiprid	135410-20-7	insecticide	X	X	0.24000
22	Acetochlor	34256-82-1	herbicide			
23	Acetochlorsulfonsäure	187022-11-3	metabolite			
24	Acetochlorsäure	194992-44-4	metabolite			
25	Acifluorfen	50594-66-6	herbicide			
26	Aclonifen	74070-46-5	herbicide	X	X	1.06000
27	Acrinathrin	101007-06-1	insecticide		X	
28	Alachlor	15972 - 60 - 8	herbicide			
29	Aldicarb	116-06-3	insecticide			
30	Aldrin	309-00-2	insecticide			
31	${ m Ametoctradin}$	865318-97-4	fungicide	X	X	
32	Ametryn	834-12-8	herbicide			
33	Amidosulfuron	120923 - 37 - 7	herbicide	X	X	
34	${ m Aminopyralid}$	150114-71-9	herbicide	X	X	
35	Amisulbrom	348635 - 87 - 0	$\operatorname{fungicide}$	X	X	
36	Anthranilsäureisopropyla	mid30391-89-0	${ m metabolite}$			
37	Atraton	1610 - 17 - 9	herbicide			
38	$\operatorname{Atrazin}$	1912-24-9	herbicide			
39	Atrazin, 2-Hydroxy	2163-68-0	${ m metabolite}$			
40	Avermectin B1a	71751-41-2	insecticide	X	X	
41	Azadirachtin (Neem)	11141 - 17 - 6	insecticide	X	X	
42	${ m Azinphos-ethyl}$	2642 - 71 - 9	insecticide			
43	${ m Azinphos-methyl}$	86-50-0	insecticide			
44	Aziprotryn	4658 - 28 - 0	herbicide			
45	${ m Azoxystrobin}$	131860-33-8	fungicide	X	X	0.55000
46	Azoxystrobin-CA		${ m metabolite}$			
47	Beflubutamid	113614 - 08 - 7	herbicide	X	X	
48	Benalaxyl	71626 - 11 - 4	$_{ m fungicide}$	X	X	20.00000
49	$\operatorname{Benazolin}$	3813-05-6	herbicide			
50	Bensulfuron-methyl	83055-99-6	herbicide		X	
51	Bentazon	25057-89-0	herbicide	X	X	535.00000
52	Benthiavalicarb	413615 - 35 - 7	fungicide	X	X	
53	Benzoesäure	65-85-0	fungicide	X	X	
54	Betacypermethrin	65731 - 84 - 2	$\overline{\mathrm{insecticide}}$		X	
55	Bifenazate	149877-41-8	insecticide	X	X	
56	Bifenox	42576-02-3	herbicide	X	X	
57	Bifenthrin	82657-04-3	insecticide		x	0.00050
58	Bixafen	581809-46-3	fungicide	X	x	0.46000
59	Boscalid	188425-85-6	fungicide	X	X	12.50000
60	Bromacil	314-40-9	herbicide			

61	Bromadiolon	28772-56-7	other		X	
62	Bromocyclen	1715-40-8	insecticide			
63	Bromoxynil	1689-84-5	herbicide	X	X	3.30000
64	Bupirimat	41483-43-6	fungicide		X	
65	Buturon	3766-60-7	herbicide			
66	Captan	133-06-2	fungicide	X	X	5.00000
67	Carbendazim	10605-21-7	fungicide			0.15000
68	Carbetamid	16118-49-3	herbicide		X	
69	Carbofuran	1563-66-2	insecticide			
70	Carboxin	5234-68-4	fungicide		X	
71	Carfentrazone-ethyl	128639-02-1	herbicide	X	X	0.31000
72	Chloramben	133-90-4	herbicide			
73	Chlorantraniliprole	500008-45-7	insecticide	X	X	0.35500
74	Chlorbromuron	13360-45-7	herbicide			
75	Chlordan	57-74-9	insecticide			
76	Chlorfenac	85-34-7	herbicide			
77	Chlorfenvinphos	470-90-6	insecticide			
78	Chlorfluazuron	71422-67-8	insecticide			
79	$\operatorname{Chloridazon}$	1698-60-8	herbicide	X	X	56.00000
80	Chlormequat	7003-89-6	other	X	X	
81	Chloroxuron	1982-47-4	herbicide			
82	Chlorpropham	101-21-3	herbicide	X	X	
83	Chlorpyrifos	2921-88-2	insecticide		X	0.00050
84	Chlorpyriphos methyl	5598-13-0	insecticide		X	
85	Chlorsulfuron	64902 - 72 - 3	herbicide			
86	Chlorthalonil	1897-45-6	fungicide	X	X	
87	Chlorthalonil-SA		$\operatorname{metabolite}$			
88	$\operatorname{Chlortoluron}$	15545-48-9	herbicide	X	X	2.30000
89	Cinidon-ethyl	142891-20-1	herbicide			
90	Clethodim	99129-21-2	herbicide	X	X	
91	Clodinafop	114420-56-3	herbicide	X	X	
92	Clodinafop-propargyl	105512-06-9	herbicide			
93	Clofentezin	74115-24-5	insecticide		X	
94	Clomazon	81777-89-1	herbicide	X	X	5.70000
95	Clopyralid	1702 - 17 - 6	herbicide	X	X	1080.00000
96	Cloquintocet-mexyl	99607-70-2	other		X	
97	Clothianidin	210880-92-5	insecticide	X	X	0.00700
98	Codlemone (Codlelure)	33956-49-9	other	X	X	
99	Coumaphos	56 - 72 - 4	insecticide			
100	Crimidin	535-89-7	other			
101	Cyanazin	21725 - 46 - 2	herbicide			
102	Cyazofamid	120116-88-3	fungicide	X	X	
103	Cyclanilide	113136-77-9	other			
104	Cycloat	1134-23-2	herbicide			

105	Cycloxidim	101205-02-1	herbicide	X	X	
106	Cyflufenamid	180409-60-3	fungicide	X	X	
107	Cyfluthrin (Summe Iso-	68359-37-5	insecticide			
	mere)					
108	Cyhalothrin (Summe	91465 - 08 - 6	insecticide	X	X	
	Isomere)					
109	Cymoxanil	57966-95-7	fungicide	X	X	4.40000
110	Cypermetryn	52315 - 07 - 8	in secticide	X	X	0.00100
111	Cyproconazol	94361 - 06 - 5	fungicide	X	X	
112	Cyprodinil	121552 - 61 - 2	$_{ m fungicide}$	X	X	0.75000
113	Cyromazin	66215 - 27 - 8	in secticide		X	
114	Daminozid	1596 - 84 - 5	other	X	X	
115	Deiquat	2764 - 72 - 9	herbicide	X	X	
116	${ m Deltamethrin}$	52918-63-5	in secticide	X	X	
117	Demeton-O	298-03-3	in secticide			
118	Demeton-S	126-75-0	in secticide			
119	Demeton-S-methyl	919-86-8	in secticide			
120	Demeton-S-	17040 - 19 - 6	in secticide			
	${ m methyl sulfon}$					
121	Desaminometribuzin	35045 - 02 - 4	${ m metabolite}$			
122	Desethyl-2-	66753 - 06 - 8	${ m metabolite}$			
	${ m hydroxyterbuthylazin}$					
123	${\bf Desethyl atrazin}$	6190 - 65 - 4	${ m metabolite}$			
124	${\bf Desethyl sebuthyl azin}$	37019 - 18 - 4	${ m metabolite}$			
125	${\bf Desethyl simazin}$	6190 - 65 - 4	${ m metabolite}$			
126	${\bf Desethylter buthylazin}$	30125 - 63 - 4	${ m metabolite}$			
127	Desisopropylatrazin	1007 - 28 - 9	${ m metabolite}$			
128	$\operatorname{Desmedipham}$	13684 - 56 - 5	herbicide	X	X	
129	${\bf Desmethyl diuron}$	3567 - 62 - 2	${ m metabolite}$			
130	${\bf Desmethylisoproturon}$	34123-57-4	${ m metabolite}$			
131	Desmetryn	1014-69-3	herbicide			
132	Desphenyl-Chloridazon	6339 - 19 - 1	${ m metabolite}$			
133	Diazinon	333-41-5	insecticide			
134	Dicamba	1918-00-9	herbicide	X	X	180.00000
135	Dichlobenil	1194-65-6	herbicide			
136	Dichlofluanid	1085 - 98 - 9	$\operatorname{fungicide}$			
137	Dichlorprop	120 - 36 - 5	herbicide			
138	Dichlorprop-P	15165-67-0	herbicide	X	X	
139	Dichlorvos	62-73-7	in secticide			
140	Diclofop	40843 - 25 - 2	herbicide		X	
141	Dicofol	115 - 32 - 2	in secticide			
142	Dieldrin	60-57-1	in secticide			
143	Difenacoum	56073-07-5	other		X	
144	Difenoconazol	119446-68-3	$\operatorname{fungicide}$	X	X	0.36000

145	Diflubenzuron	35367-38-5	insecticide		X	
146	Diflufenican	83164-33-4	herbicide	X	X	0.02500
147	Dimefuron	34205 - 21 - 5	herbicide			0.83000
148	Dimethachlor	50563-36-5	herbicide	X	X	3.50000
149	Dimethachlor-CA		${ m metabolite}$			
150	Dimethachlorsulfonsäure		${ m metabolite}$			
151	Dimethachlorsäure		${ m metabolite}$			
152	Dimethenamid	87674-68-8	herbicide			
153	Dimethenamid-CA		$_{ m metabolite}$			
154	Dimethenamid-P	163515-14-8	herbicide	X	X	1.35000
155	Dimethenamid-SA		$_{ m metabolite}$			
156	Dimethenamidsulfonsäure		${ m metabolite}$			
157	Dimethoat	60-51-5	insecticide	X	X	4.00000
158	Dimethomorph	110488-70-5	fungicide	X	X	5.60000
159	Dimoxystrobin	149961 - 52 - 4	fungicide	X	X	0.03160
160	Diniconazol	83657-24-3	fungicide			
161	Dinoseb	88-85-7	herbicide			
162	Dinotefuran	165252 - 70 - 0	insecticide			
163	Dinoterb	1420 - 07 - 1	herbicide			
164	Disulfoton	298-04-4	insecticide			
165	Dithianon	3347 - 22 - 6	fungicide	X	X	0.78000
166	Diuron	330 - 54 - 1	herbicide		X	0.79000
167	Dodin	2439 - 10 - 3	fungicide	X	X	5.33000
168	Endosulfan, alpha	959-98-8	insecticide			
169	Endosulfan, beta	33213-65-9	insecticide			
170	Endosulfansulfat	1031-07-8	${ m metabolite}$			
171	Endrin	72-20-8	insecticide			
172	Epoxiconazol	133855-98-8	fungicide	X	X	0.53750
173	Esfenvalerat	66230-04-4	insecticide	X	X	
174	Etaconazol	60207 - 93 - 4	fungicide			
175	Ethidimuron	30043-49-3	herbicide			
176	Ethirimol	23947-60-6	fungicide			
177	Ethofenprox	80844-07-1	insecticide	X	X	
178	Ethofumes at	26225 - 79 - 6	herbicide	X	X	24.00000
179	Etrimfos	38260 - 54 - 7	insecticide			
180	Famoxadone	131807-57-3	fungicide	X	X	
181	Fenamidon	161326 - 34 - 7	$_{ m fungicide}$	X	X	
182	Fenamiphos	22224-92-6	insecticide		X	
183	Fenarimol	60168-88-9	fungicide			
184	Fenazaquin	120928-09-8	insecticide	X	X	
185	Fenhexamid	126833-17-8	$_{ m fungicide}$	X	X	10.10000
186	Fenitrothion	122 - 14 - 5	in sectic ide			
187	Fenoprop	93-72-1	herbicide			
188	Fenoxaprop	95617-09-7	herbicide			

189	Fenoxaprop-p	113158-40-0	herbicide	X	X	
190	Fenoxaprop-p-ethyl	71283-80-2	herbicide			
191	Fenoxycarb	72490-01-8	insecticide		X	
192	Fenpropidin	67306-00-7	fungicide	X	X	
193	Fenpropimorph	67564-91-4	fungicide	X	X	0.19500
194	Fenpyroximat	134098-61-6	insecticide	X	X	
195	Fenthion	55 - 38 - 9	insecticide			
196	Fenuron	101-42-8	herbicide			
197	Fipronil	120068 - 37 - 3	insecticide		X	0.00077
198	Flamprop	58667 - 63 - 3	herbicide			
199	Flazasulfuron	104040-78-0	herbicide	X	X	
200	Flonicamid	158062 - 67 - 0	insecticide	X	X	310.00000
201	Florasulam	145701 - 23 - 1	herbicide	X	X	
202	Fluazifop	69335-91-7	herbicide			146.00000
203	Fluazifop-P	83066-88-0	herbicide	X	X	146.00000
204	Fluazifop-P-butyl	79241-46-6	herbicide			7.70000
205	Fluazifop-butyl	69806-50-4	herbicide			7.70000
206	Fluazinam	79622-59-6	fungicide	X	X	0.26000
207	Fluchloralin	33245-39-5	herbicide			
208	Fludioxonil	131341-86-1	fungicide	X	X	0.50000
209	Flufenacet	142459-58-3	herbicide	X	X	2.40000
210	Flufenacet-SA		metabolite			
211	Flufenoxuron	101463-69-8	insecticide			
212	Flumioxazin	103361-09-7	herbicide	X	X	
213	Fluometuron	2164-17-2	herbicide		X	
214	Fluopicolide	239110-15-7	fungicide	X	X	
215	Fluopyram	658066-35-4	fungicide	X	X	5.12000
216	Fluoxastrobin	361377-29-9	fungicide	X	X	
217	Flupyrsulfuron	150315-10-9	herbicide	X	X	
218	Fluquinconazole	136426-54-5	fungicide	X	X	0.80000
219	Flurochloridon	61213-25-0	herbicide		X	
220	Fluroxypyr	69377-81-7	herbicide	X	X	16.00000
221	Fluroxypyr-	81406-37-3	herbicide			
	methylheptyl	02100 0, 0	11010101010			
222	Flurtamone	96525-23-4	herbicide	X	X	0.99000
223	Flusilazol	85509-19-9	fungicide	11	21	1.10000
224	Flutolanil	66332-96-5	fungicide	X	X	1110000
225	Flutriafol	76674-21-0	fungicide	21	X	
226	Fluxapyroxad	907204-31-3	fungicide	X	X	
227	Folpet	133-07-3	fungicide	X	X	
228	Foramsulfuron	173159-57-4	herbicide	X	X	0.09500
$\frac{220}{229}$	Fosetyl	15845-66-6	fungicide	X	X	0.0000
$\frac{229}{230}$	Fosthiazat	98886-44-3	other	X	X	
$\frac{230}{231}$	Fuberidazol	3878-19-1	fungicide			
7 O T	r abelidazoi	9010-19-1	rangidae	X	X	

232	Furalaxyl	57646-30-7	fungicide			
233	Furmecyclox	60568-05-0	fungicide			
234	Glufosinat	51276-47-2	herbicide	X	X	
235	Glyphosate	1071-83-6	herbicide	X	X	100.00000
236	HCH, gamma (Lindan)	58-89-9	insecticide			
237	Haloxyfop	69806-34-4	herbicide			
238	Haloxyfop-P	95977-29-0	herbicide	X	X	
239	Haloxyfop-ethoxyethyl	87237-48-7	herbicide			
$\frac{240}{240}$	Heptachlor	76-44-8	insecticide			
241	Heptachlorepoxid	1024-57-3	metabolite			
242	Heptenophos	23560-59-0	insecticide			
243	Hexachlorbenzen	118-74-1	fungicide			
244	Hexachlorophen	70-30-4	other			
245	Hexaconazol	79983-71-4	fungicide			
246	Hexaflumuron	86479-06-3	insecticide			
247	Hexazinon	51235-04-2	herbicide			
248	Hexythiazox	78587-05-0	insecticide	X	X	
249	Hymexazol	10004-44-1	fungicide	X	X	
250	Icaridinsäure		$_{ m metabolite}$			
251	Imazalil	35554-44-0	fungicide	X	X	
252	Imazamox	114311-32-9	herbicide	X	X	
253	Imazapic	104098-48-8	herbicide			
254	Imazaquin	81335-37-7	herbicide		X	
255	Imazethapyr	81335-77-5	herbicide			
256	Imazosulfuron	122548-33-8	herbicide	X	X	
257	Imidacloprid	138261-41-3	insecticide	X	X	0.00900
258	Indoxacarb	173584-44-6	insecticide	X	X	
259	Iodosulfuron	185119-76-0	herbicide	X	X	0.07900
260	Iodosulfuron-methyl	144550-06-1	herbicide			
261	Iodosulfuron-methyl-	144550-36-7	herbicide			
	sodium					
262	Ioxynil	1689-83-4	herbicide	X		2.70000
263	Iprodion	36734-19-7	fungicide	X	X	
264	Iprovalicarb	140923-17-7	fungicide	X	X	189.00000
265	Isodrin	465 - 73 - 6	insecticide			
266	Isophenphos	25311-71-1	insecticide			
267	Isoproturon	34123-59-6	herbicide	X	x	1.30000
268	Isopyrazam	881685-58-1	fungicide	X	X	
269	Isoxaben	82558-50-7	herbicide	X	X	
270	Isoxaflutole	141112-29-0	herbicide	X	X	
271	Karbutylat	4849-32-5	herbicide			
272	Kresoxim-methyl	143390-89-0	fungicide	X	X	1.00000
273	Kresoximsäure		$\operatorname{metabolite}$			
274	Lenacil	2164-08-1	herbicide	X	X	0.65000

275	Linuron	330-55-2	herbicide		X	
276	MCPA	94-74-6	herbicide	X	X	9.00000
277	MCPB	94-81-5	herbicide		X	
278	Malathion	121-75-5	insecticide		X	
279	Mancozeb	8018-01-7	fungicide	X	X	0.21900
280	Mandipropamid	374726-62-2	fungicide	X	X	7.60000
281	Maneb	12427-38-2	fungicide	X	X	
282	Mecoprop	93-65-2	herbicide		X	160.00000
283	Mefenpyr-diethyl	135591-00-3	other	X		
284	Mepanipyrim	110235-47-7	fungicide	X	X	
285	Mepiquat	15302-91-7	other	X	X	
286	Mepronil	55814-41-0	fungicide			
287	Meptyldinocap	131-72-6	fungicide		X	
288	Mesosulfuron	400852-66-6	herbicide	X	X	
289	Mesotrion	104206-82-8	herbicide	X	X	
290	Metaflumizone	139968-49-3	insecticide	X	X	
291	Metalaxyl	57837-19-1	fungicide		X	46.00000
292	Metalaxyl-CA	75596-99-5	${ m metabolite}$			
293	Metalaxyl-CA2	104390-56-9	${ m metabolite}$			
294	Metalaxyl-M	70630 - 17 - 0	fungicide	X	X	46.00000
295	Metaldehyd	108-62-3	other	X	X	
296	Metamitron	41394 - 05 - 2	herbicide	X	X	38.00000
297	Metamitron-Desamino	36993-94-9	${ m metabolite}$			
298	Metazachlor	67129 - 08 - 2	herbicide	X	X	0.88000
299	Metazachlordicarbonsäur	e	${ m metabolite}$			
300	${ m Metazachlor sulfons \ddot{a}ure}$	172960-62-2	${ m metabolite}$			
301	Metazachlorsäure	1231244-60-2	${ m metabolite}$			
302	Metconazol	125116 - 23 - 6	$_{ m fungicide}$	X	X	
303	${ m Methabenzthiazuron}$	18691 - 97 - 9	herbicide			
304	Methamidophos	10265 - 92 - 6	in sectic ide			2.60000
305	Methidathion	950-37-8	insecticide			
306	Methiocarb	2032 - 65 - 7	insecticide	X	X	0.01000
307	Methobromuron	3060-89-7	herbicide		X	2.00000
308	Methomyl	16752 - 77 - 5	insecticide		X	
309	Methoprotryn	841-06-5	herbicide			
310	Methoxychlor	72 - 43 - 5	insecticide			
311	Methoxyfenozid	161050-58-4	insecticide	X	X	
312	Methyldesphenyl- Chloridazon	17254-80-7	${ m metabolite}$			
313	Metiram	9006-42-2	fungicide	X	X	
314	Metolachlor	51218-45-2	herbicide			
315	Metolachlorsulfonsäure	171118-09-5	${ m metabolite}$			
316	Metolachlorsäure	152019-73-3	${ m metabolite}$			
317	Metosulam	139528-85-1	herbicide	X	X	

319 Metrafenon 220899-03-6 fungicide x x 320 Metribuzin 21087-64-9 herbicide x x	
320 Metribuzin 21087-64-9 herbicide v v	
GEO MICHIGIAN A A	0.58400
321 Metsulfuron 79510-48-8 herbicide x x	
322 Metsulfuron-methyl 74223-64-6 herbicide	
323 Mevinphos 7786-34-7 insecticide	
324 Milbemectin 51596-11-3 insecticide x x	
325 Mirex 2385-85-5 insecticide	
326 Monolinuron 1746-81-2 herbicide	
327 Monuron 150-68-5 herbicide	
328 Myclobutanil 88671-89-0 fungicide x x	2.40000
	6.70000
330 Neburon 555-37-3 herbicide	
331 Nicosulfuron 111991-09-4 herbicide x x	0.08500
332 Nitenpyram 120738-89-8 insecticide	
333 Nitrofen 1836-75-5 herbicide	
334 Norflurazon 27314-13-2 herbicide	
335 Omethoat 1113-02-6 insecticide	
336 Orysastrobin 248593-16-0 fungicide	
337 Oxadiazon 19666-30-9 herbicide x	
338 Oxadixyl 77732-09-3 fungicide	
339 Oxamyl 23135-22-0 insecticide x	
340 Oxydemeton-methyl 301-12-2 insecticide	1.10000
341 Paclobutrazol 76738-62-0 other x x	
342 Parathion-ethyl 56-38-2 insecticide	
343 Parathion-methyl 298-00-0 insecticide	
344 Pelargonsäure 112-05-0 herbicide x x	
345 Penconazol 66246-88-6 fungicide x x	3.20000
346 Pencycuron 66063-05-6 fungicide x x	
347 Pendimethalin 40487-42-1 herbicide x x	0.63000
348 Penflufen 494793-67-8 fungicide x	
349 Penoxsulam 219714-96-2 herbicide x x	
350 Permethrin 52645-53-1 insecticide	
351 Pethoxamid 106700-29-2 herbicide x x	1.77200
352 Phenmedipham 13684-63-4 herbicide x x	
353 Phoxim 14816-18-3 insecticide	0.00700
354 Picloram 1918-02-1 herbicide x x	
355 Picolinafen 137641-05-5 herbicide x x	0.03600
356 Picoxystrobin 117428-22-5 fungicide x x	0.60000
357 Pinoxaden 243973-20-8 herbicide x	
358 Pirimicarb $23103-98-2$ insecticide x x	0.09000
359 Pirimicarb-desmethyl 30614-22-3 metabolite	
360 Pirimiphos-ethyl 23505-41-1 insecticide	
361 Pirimiphos-methyl 29232-93-7 insecticide x x	

362	Primisulfuron-methyl	86209-51-0	herbicide			
363	Prochloraz	67747 - 09 - 5	fungicide	X	X	5.00000
364	Procymidon	32809 - 16 - 8	fungicide			
365	Profoxydim	139001 - 49 - 3	herbicide		X	
366	Prohexadion	88805-35-0	other	X	X	
367	Prometryn	7287 - 19 - 6	herbicide			
368	Propachlor	1918 - 16 - 7	herbicide			
369	Propamocarb	24579 - 73 - 5	fungicide	X	X	
370	Propanil	709-98-8	herbicide			
371	Propaquizafop	111479 - 05 - 1	herbicide	X	X	
372	Propazin	139 - 40 - 2	herbicide			
373	Propetamphos	31218-83-4	insecticide			
374	Propham	122 - 42 - 9	herbicide			
375	Propiconazol	60207 - 90 - 1	$_{ m fungicide}$	X	X	2.00000
376	Propoxur	114 - 26 - 1	insecticide			
377	Propoxycarbazone	145026 - 81 - 9	herbicide	X	X	
378	Propyzamid	23950 - 58 - 5	herbicide	X	X	34.00000
379	Proquinazid	189278 - 12 - 4	fungicide	X	X	
380	Prosulfocarb	52888-80-9	herbicide	X	X	3.80000
381	Prosulfuron	94125 - 34 - 5	herbicide	X	X	
382	Prothioconazol	178928-70-6	fungicide	X	X	1.71000
383	Prothioconazol-desthio	120983 - 64 - 4	${ m metabolite}$			
384	Pymetrozin	123312 - 89 - 0	insecticide	X	X	
385	Pyraclostrobin	175013 - 18 - 0	fungicide	X	X	
386	Pyraflufen	129630 - 17 - 7	herbicide	X	X	
387	Pyrazophos	13457 - 18 - 6	fungicide			
388	Pyrethrum	8003-34-7	insecticide	X	X	0.01400
389	Pyridaben	96489 - 71 - 3	insecticide		X	
390	Pyridat	55512 - 33 - 9	herbicide	X	X	
391	Pyrifenox	88283-41-4	fungicide			
392	Pyrimethanil	53112 - 28 - 0	fungicide	X	X	8.00000
393	Pyroxsulam	422556 - 08 - 9	herbicide	X	X	
394	Quinalphos	13593 - 03 - 8	insecticide			
395	Quinmerac	90717 - 03 - 6	herbicide	X	X	316.00000
396	Quinoclamin	2797 - 51 - 5	herbicide	X	X	
397	Quinoxyfen (5,7-	124495 - 18 - 7	$_{ m fungicide}$	X	X	
	dichloro-4-(p-					
	fluorophenoxy)quinoline)					
398	Quintozen	82-68-8	fungicide			
399	Quizalofop	76578 - 12 - 6	herbicide			
400	Quizalofop-ethyl	76578 - 14 - 8	herbicide			
401	Rimsulfuron	122931 - 48 - 0	herbicide	X	X	0.46000
402	Saflufenacil	372137 - 35 - 4	herbicide			
403	Sebuthylazin	7286-69-3	herbicide			

404	Secbumeton	26259-45-0	herbicide			
405	$\operatorname{Silthiofam}$	175217-20-6	fungicide	X	X	
406	Simazin	122-34-9	herbicide			
407	Simazin, 2-Hydroxy	2599 - 11 - 3	${ m metabolite}$			
408	Spinosad	168316-95-8	insecticide	X	X	0.06200
409	Spirodiclofen	148477-71-8	insecticide	X	X	
410	Spiromesifen	283594-90-1	insecticide		X	
411	Spiroxamin	118134-30-8	fungicide	X	X	0.13000
412	Sulcotrion	99105-77-8	herbicide	X	X	
413	Sulfosulfuron	141776-32-1	herbicide		X	
414	Sulfurylfluorid	2699-79-8	insecticide	X	X	
415	Tebuconazol	107534-96-3	fungicide	X	X	0.57800
416	Tebufenozid	112410-23-8	$\frac{1}{2}$ insecticide	X	X	
417	Tebufenpyrad	119168-77-3	insecticide	X	X	
418	Tebutam	35256-85-0	herbicide			
419	Teflubenzuron	83121-18-0	insecticide		X	
420	Tefluthrin	79538-32-2	insecticide	X	X	
421	Telodrin	297-78-9	insecticide			
422	Tembotrione	335104-84-2	herbicide	X	X	
423	Tepraloxydim	149979-41-9	herbicide	X	X	
424	Terbumeton	33693-04-8	herbicide			
425	Terbuthylazin	5915-41-3	herbicide	X	X	1.20000
426	Terbutryn	886-50-0	herbicide			
427	Terbutylazin-Metabolit	309923-18-0	${ m metabolite}$			
	CGA 324007					
428	Terbutylazin-Metabolit		${ m metabolite}$			
	SYN 545666					
429	Tetraconazol	112281-77-3	fungicide	X	X	
430	Thiabendazol	148-79-8	fungicide	X	X	
431	Thiacloprid	111988-49-9	$\inf_{insecticide}$	X	X	0.00400
432	Thiacloprid-SA		${ m metabolite}$			
433	Thiamethoxam	153719 - 23 - 4	insecticide	X	X	0.04300
434	Thiencarbazon-methyl	317815-83-1	herbicide	X	X	
435	Thifensulfuron-methyl	79277-27-3	herbicide			
436	Thifenylsulfuron	79277-67-1	herbicide	X	X	
437	Thiometon	640-15-3	insecticide			
438	Thiophanat-methyl	23564-05-8	fungicide	X	X	
439	Thiram	137-26-8	fungicide	X	X	0.11000
440	Tolclofos-methyl	57018-04-9	fungicide	X	X	
441	Tolylfluanid	731-27-1	fungicide			
442	Topramezone	210631-68-8	herbicide	X		0.90000
443	Tralkoxydim	87820-88-0	herbicide		X	
444	Triadimefon	43121-43-3	fungicide			
445	Triadimenol	55219-65-3	fungicide	X	X	3.40000

446	Triallat	2303-17-5	herbicide		X	
447	Triasulfuron	82097-50-5	herbicide	X	X	
448	Triazophos	24017 - 47 - 8	insecticide			0.03000
449	Triazoxid	72459 - 58 - 6	fungicide	X	X	
450	Tribenuron	106040 - 48 - 6	herbicide	X	X	
451	Tribenuron-methyl	101200 - 48 - 0	herbicide			
452	Trichlorfon	52-68-6	insecticide			
453	Triclopyr	55335-06-3	herbicide	X	X	
454	Trifloxystrobin	141517 - 21 - 7	fungicide	X	X	0.08620
455	Trifloxystrobin-CA2		${ m metabolite}$			
456	Triflumizol	99387-89-0	fungicide		X	
457	Triflumuron	64628 - 44 - 0	insecticide		X	
458	Trifluralin	1582 - 09 - 8	herbicide			
459	Triflusulfuron	135990 - 29 - 3	herbicide	X	X	
460	Triforin	26644 - 46 - 2	fungicide			
461	Trinexapac-ethyl	95266 - 40 - 3	other	X	X	
462	Triticonazol	131983 - 72 - 7	fungicide	X	X	
463	Tritosulfuron	142469 - 14 - 5	herbicide	X	X	
464	Valifenalate	283159-90-0	$\operatorname{fungicide}$	X	X	
465	Vinclozolin	50471 - 44 - 8	fungicide			
466	Warfarin	81-81-2	other			
467	Zoxamid	156052 - 68 - 5	fungicide	X	X	
468	alpha-Cypermethrin	67375 - 30 - 8	insecticide	X	X	
469	cis-Chlordan	5103 - 71 - 9	insecticide			
470	gamma-Cyhalothrin	76703 - 62 - 3	insecticide	X	X	
471	$_{ m o,p ext{-}DDE}$	3424-82-6	${ m metabolite}$			
472	$_{ m o,p ext{-}DDT}$	789-02-6	insecticide			
473	oxi-Chlordan	27304 - 13 - 8	${ m metabolite}$			
474	p,p-DDD (p,p TDE)	72 - 54 - 8	insecticide			
475	$_{\mathrm{p,p-DDE}}$	72 - 55 - 9	${ m metabolite}$			
476	$_{\mathrm{p,p-DDT}}$	50-29-3	insecticide			
477	tau-Fluvalinat	102851-06-9	insecticide	X	X	0.03300
478	trans-Chlordan	5103-74-2	in sectic ide			

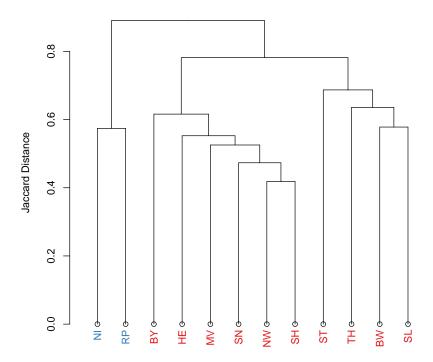


Figure S3: Complete Linkage Cluster Dendrogram of Jaccard Similarity of analysed compound spectra between federal states. Abbreviations of state names according to ISO 3166-2:DE (see also Table S1).

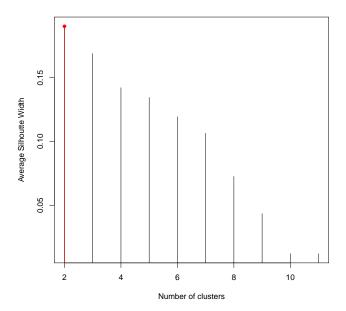


Figure S4: Average silhouette width for different cluster sizes of complete linkage clustering of jaccard similarity of analysed compound spectra between federal states. Two clusters showed maximum silhouette width.

3 Thresholds for agricultural land use and catchment size

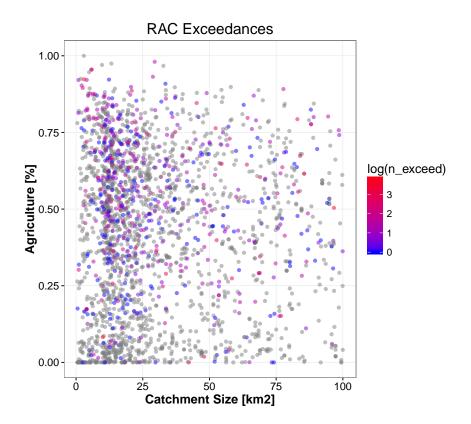


Figure S5: Raw data used for the model in equation 2 and Figure 3 of the main article. Color codes the number of RAC exceedances (on a log-scale). Grey points denote sites without any exceedance.

4 Effect of precipitation and season on RQ

Table S3: 23 pesticides for which we modelled the relationship between RQ and precipitation and seasonality, respectively. Order is the same as in Figure 5 of the main text. See Table S4 for model coefficients.

	Name	CAS	Group	%>LOQ	${ m no.} > { m LOQ}$	total no.
1	Azoxystrobin	131860-33-8	$_{ m fungicide}$	9.58	644	6723
2	$\operatorname{Bentazon}$	25057 - 89 - 0	herbicide	19.43	2313	11905
3	Boscalid	188425-85-6	$_{ m fungicide}$	23.00	2175	9455
4	Carbendazim	10605 - 21 - 7	fungicide	16.10	582	3615
5	Chlorpyrifos	2921-88-2	insecticide	6.17	865	14026
6	Clothianidin	210880-92-5	in secticide	6.30	141	2237
7	Diflufenican	83164-33-4	herbicide	12.63	1867	14784
8	Dimoxystrobin	149961 - 52 - 4	$_{ m fungicide}$	6.83	216	3164
9	Diuron	330 - 54 - 1	herbicide	12.07	2138	17708
10	Ethofumes at	26225 - 79 - 6	herbicide	5.10	998	19552
11	Flufenacet	142459-58-3	herbicide	5.97	772	12923
12	$\operatorname{Glyphosate}$	1071 - 83 - 6	herbicide	40.73	1389	3410
13	Imidacloprid	138261-41-3	in secticide	5.88	176	2992
14	Isoproturon	34123 - 59 - 6	herbicide	21.84	3984	18239
15	MCPA	94 - 74 - 6	herbicide	12.81	1567	12237
16	Mecoprop	93 - 65 - 2	herbicide	12.21	1463	11984
17	Metazachlor	67129 - 08 - 2	herbicide	9.23	1930	20907
18	Nicosulfuron	111991-09-4	herbicide	5.33	263	4934
19	Propiconazol	60207 - 90 - 1	$_{ m fungicide}$	5.67	772	13622
20	$\operatorname{Quinmerac}$	90717-03-6	herbicide	13.46	939	6974
21	Tebuconazol	107534 - 96 - 3	$_{ m fungicide}$	6.08	968	15924
22	Terbuthylazin	5915-41-3	herbicide	14.59	3142	21540

Table S4: Coefficients and CI from per compound models. Bold values denote coefficients where the CI for precipitation encompasses zero. Coefficients are on the link scale (log for μ and logit for ν).

C	Compound	effect	$log\ precip_0$	$log\ precip_{-1}$	Quarter 1	Quarter 2	Quarter 3	Quarter 4

1	A		0.00	0.04	2.20	2.00	2.10	2.47
1	Azoxystrobin	μ	0.23 (0.15 - 0.31)	0.04 (-0.03 - 0.12)	-3.39 (-3.563.22)	-3.02 (-3.142.89)	-3.16 (-3.293.03)	-3.47 (-3.633.3)
2	Bentazon	μ	-0.03	0.02	-9.74	-9.25	-9.42	-9.74
		•	(-0.07 - 0)	(-0.02 - 0.05)	(-9.819.67)	(-9.319.2)	(-9.489.36)	(-9.819.68)
3	Boscalid	μ	0.06	0.1	-6.72	-6.42	-6.51	-6.58
			(0.02 - 0.1)	(0.06 - 0.13)	(-6.796.64)	(-6.496.36)	(-6.586.45)	(-6.656.5)
4	Carbendazim	μ	-0.1	0.16	-2.42	-1.95	-2.11	-2.32
5	Chlannynifaa		(-0.160.03) 0.08	(0.09 - 0.22) -0.03	(-2.582.27) 0.95	(-2.051.84) 1.1	(-2.222) 1	(-2.462.18) 1.05
Э	Chlorpyrifos	μ	(0.04 - 0.13)	(-0.08 - 0.01)	(0.88 - 1.03)	(1.03 - 1.17)	(0.93 - 1.08)	(0.96 - 1.14)
6	Clothianidin	μ	0.08	-0.1	0.94	0.67	1.02	1.55
		μ.	(-0.04 - 0.19)	(-0.21 - 0.02)	(0.77 - 1.12)	(0.49 - 0.84)	(0.8 - 1.25)	(1.32 - 1.78)
7	Diflufenican	μ	-0.02	0.05	-0.56	-1.01	-1.08	-0.71
			(-0.06 - 0.02)	(0.02 - 0.09)	(-0.620.5)	(-1.070.94)	(-1.161)	(-0.770.65)
8	Dimethenamid	μ	-0.1	0.05	-4.01	-3.79	-3.72	-4.02
			(-0.170.03)	(-0.01 - 0.12)	(-4.163.86)	(-3.893.68)	(-3.843.61)	(-4.143.9)
9	Dimoxystrobin	μ	0.35	0.02	-1.17	-0.42	-0.07	-0.02
10	Diuron		(0.2 - 0.5)	(-0.15 - 0.19) 0.07	(-1.440.89) -2.72	(-0.640.2) -2.43	(-0.39 - 0.25) -2.48	(-0.35 - 0.31) -2.64
10	Diulon	μ	(-0.03 - 0.03)	(0.04 - 0.1)	(-2.832.61)	(-2.472.39)	(-2.532.44)	(-2.712.58)
11	Ethofumesat	μ	0.12	0.01	-6.11	-5.49	-6.18	-6.1
		,	(0.06 - 0.17)	(-0.05 - 0.06)	(-6.275.96)	(-5.565.42)	(-6.296.08)	(-6.245.95)
12	Flufenacet	μ	0.03	0.05	-3.71	-3.7	-3.29	-3.63
			(-0.02 - 0.08)	(0.01 - 0.1)	(-3.793.62)	(-3.813.59)	(-3.443.15)	(-3.683.57)
13	Glyphosate	μ	-0.04	0.14	-6.3	-6.08	-5.73	-6.11
1.4	T '1 1 '1		(-0.09 - 0.01)	(0.09 - 0.19)	(-6.466.13)	(-6.166)	(-5.85.66)	(-6.216.01)
14	Imidacloprid	μ	0 (-0.08 - 0.09)	-0.01 (-0.09 - 0.07)	0.61 $(0.33 - 0.88)$	1.15 $(1.02 - 1.27)$	1.4 $(1.28 - 1.53)$	1.24 $(1.06 - 1.42)$
15	Isoproturon	μ	0.02	(-0.09 - 0.07) 0.21	-3.29	-3.01	-3.43	-2.79
10	Isopiotaion	μ	(-0.02 - 0.05)	(0.17 - 0.24)	(-3.373.22)	(-3.062.96)	(-3.53.35)	(-2.842.73)
16	MCPA	μ	0.04	0.09	-5.07	-4.25	-4.48	-4.7
		•	(-0.01 - 0.09)	(0.04 - 0.14)	(-5.274.87)	(-4.324.19)	(-4.574.4)	(-4.814.58)
17	Mecoprop	μ	0.04	0.05	-8.36	-7.59	-7.77	-8.07
			(-0.01 - 0.09)	(0.01 - 0.1)	(-8.498.22)	(-7.657.52)	(-7.857.69)	(-8.187.97)
18	Metazachlor	μ	-0.07	0.09	-2.97	-2.94	-2.21	-2.77
19	Nicosulfuron		(-0.120.02) 0.23	(0.04 - 0.13) - 0.28	(-3.062.88) -0.98	(-3.042.85) -0.2	(-2.282.14) -0.07	(-2.842.7) -0.97
19	Nicosulluloli	μ	(0.12 - 0.34)	(-0.390.18)	(-1.220.74)	(-0.360.03)	(-0.25 - 0.11)	(-1.160.78)
20	Propiconazol	μ	0.08	0.01	-3.99	-3.63	-3.82	-3.63
	r	,	(0.02 - 0.14)	(-0.05 - 0.07)	(-4.153.83)	(-3.713.55)	(-3.913.72)	(-3.743.53)
21	$\operatorname{Quinmerac}$	μ	0.02	0.05	-9.08	-9.12	-8.46	-8.64
			(-0.05 - 0.09)	(-0.01 - 0.12)	(-9.198.96)	(-9.249)	(-8.598.33)	(-8.728.55)
22	Tebuconazol	μ	-0.01	0.09	-2.17	-1.93	-2.2	-2.15
0.9	Tonbuthylogin		(-0.06 - 0.03) 0.09	(0.04 - 0.14)	(-2.282.06) -3.65	(-21.86)	(-2.282.11) -3.25	(-2.242.06) -3.52
23	Terbuthylazin	μ	(0.06 - 0.13)	0.11 (0.08 - 0.15)	-3.733.56)	-2.78 (-2.842.73)	-3.25 (-3.33.19)	-3.52 (-3.593.44)
24	Azoxystrobin	ν	0	0.24	-3.5	-2.33	-2.14	-3.2
0.5	D		(-0.13 - 0.13)	(0.11 - 0.37)	(-3.763.25)	(-2.542.13)	(-2.361.92)	(-3.452.95)
25	Bentazon	ν	0 (-0.08 - 0.08)	0.05 (-0.03 - 0.13)	-2.26 (-2.442.09)	-1.53 (-1.651.4)	-1.88 (-2.021.74)	-2.25 (-2.42.11)
26	Boscalid	ν	-0.01	0.45	-1.99	-1.22	-1.24	-1.81
	Bosowna	-	(-0.1 - 0.08)	(0.37 - 0.54)	(-2.161.82)	(-1.361.07)	(-1.381.09)	(-1.961.65)
27	Carbendazim	ν	0.09	0.19	-2.72	-1.49	-1.26	-2.31
			(-0.04 - 0.22)	(0.06 - 0.32)	(-32.44)	(-1.691.28)	(-1.481.04)	(-2.562.06)
28	Chlorpyrifos	ν	0.11	0.1	-3.27	-2.63	-3.22	-3.42
	er		(0.01 - 0.21)	(0 - 0.19)	(-3.453.1)	(-2.792.48)	(-3.393.05)	(-3.613.23)
29	Clothianidin	ν	-0.05	0.19	-2.66	-2.58	-3.19	-3.93
			(-0.3 - 0.2)	(-0.07 - 0.44)	(-3.062.26)	(-2.972.19)	(-3.692.69)	(-4.463.41)

30	Diflufenican	ν	0.06	0.26	-1.89	-2.45	-3.14	-2.09
			(-0.02 - 0.14)	(0.17 - 0.34)	(-2.031.75)	(-2.592.31)	(-3.32.98)	(-2.221.95)
31	Dimethenamid	ν	-0.04	0.22	-3.44	-2.7	-2.79	-2.99
			(-0.16 - 0.08)	(0.11 - 0.34)	(-3.693.2)	(-2.882.51)	(-2.982.59)	(-3.192.78)
32	Dimoxystrobin	ν	0.19	0.23	-3.37	-2.25	-3.14	-3.58
			(-0.02 - 0.41)	(0.01 - 0.46)	(-3.782.96)	(-2.581.91)	(-3.552.72)	(-4.023.15)
33	Diuron	ν	0.05	0.28	-3.88	-1.67	-1.74	-2.72
			(-0.01 - 0.12)	(0.22 - 0.35)	(-4.093.67)	(-1.761.58)	(-1.841.63)	(-2.852.6)
34	Ethofumesat	ν	0.09	0.21	-4.39	-2.23	-3.49	-4.23
			(-0.01 - 0.18)	(0.12 - 0.3)	(-4.634.16)	(-2.352.11)	(-3.663.32)	(-4.444.01)
35	Flufenacet	ν	0.16	0.59	-2.57	-3.8	-4.17	-1.76
			(0.06 - 0.27)	(0.49 - 0.69)	(-2.752.39)	(-4.013.58)	(-4.443.89)	(-1.881.64)
36	Glyphosate	ν	0.11	0.29	-1.79	-0.12	0.34	-0.53
			(0 - 0.23)	(0.18 - 0.4)	(-2.091.48)	(-0.3 - 0.05)	(0.17 - 0.51)	(-0.730.32)
37	Imidacloprid	ν	-0.01	-0.1	-4.68	-3.04	-2.83	-4.07
			(-0.26 - 0.25)	(-0.34 - 0.15)	(-5.354)	(-3.412.68)	(-3.212.45)	(-4.563.58)
38	Isoproturon	ν	0.04	0.31	-1.82	-1.19	-2.11	-0.8
			(-0.02 - 0.09)	(0.25 - 0.36)	(-1.931.7)	(-1.271.12)	(-2.222.01)	(-0.880.72)
39	MCPA	ν	-0.06	0.35	-3.79	-1.27	-1.81	-2.77
			(-0.13 - 0.02)	(0.28 - 0.42)	(-4.043.54)	(-1.371.18)	(-1.931.68)	(-2.922.62)
40	Mecoprop	ν	0.07	0.35	-3.04	-1.56	-1.89	-2.71
			(-0.01 - 0.15)	(0.27 - 0.42)	(-3.232.84)	(-1.671.45)	(-2.021.76)	(-2.862.56)
41	Metazachlor	ν	0.06	0.21	-2.81	-3.22	-2.11	-2.05
			(-0.01 - 0.13)	(0.14 - 0.27)	(-2.942.67)	(-3.363.09)	(-2.222.01)	(-2.161.95)
42	Nicosulfuron	ν	0.2	0.26	-3.87	-2.96	-2.99	-3.23
			(0.01 - 0.39)	(0.07 - 0.45)	(-4.273.48)	(-3.262.66)	(-3.32.68)	(-3.562.9)
43	Propiconazol	ν	-0.02	0.39	-4.05	-2.72	-2.88	-3.43
			(-0.13 - 0.09)	(0.29 - 0.5)	(-4.323.78)	(-2.882.57)	(-3.062.7)	(-3.633.24)
44	Quinmerac	ν	-0.03	0.32	-2.23	-2.58	-2.49	-1.2
			(-0.13 - 0.08)	(0.22 - 0.42)	(-2.432.02)	(-2.762.41)	(-2.692.29)	(-1.341.06)
45	Tebuconazol	ν	0.1	0.3	-3.41	-2.66	-2.9	-3.17
			(0.01 - 0.2)	(0.21 - 0.39)	(-3.613.2)	(-2.82.53)	(-3.062.75)	(-3.343)
46	Terbuthylazin	ν	0.06	0.28	-2.92	-1.45	-1.48	-2.47
	-		(0.01 - 0.12)	(0.22 - 0.33)	(-3.052.79)	(-1.531.37)	(-1.571.39)	(-2.582.37)

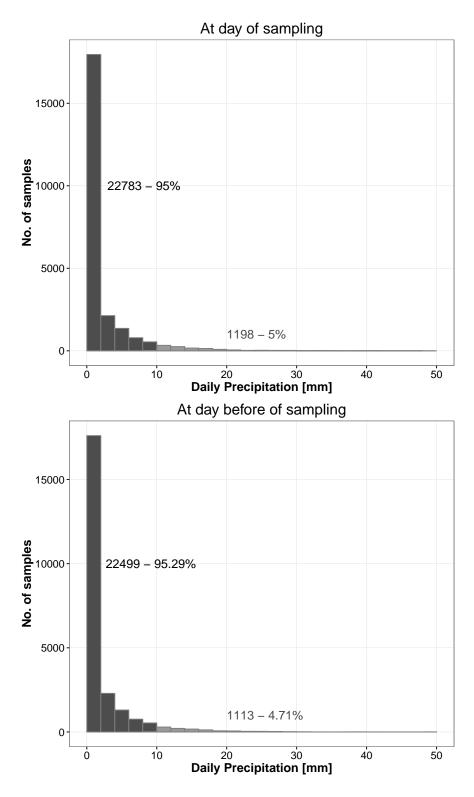


Figure S6: Distribution of precipitation at sampling occasions. top: at sampling date. bottom: at day before sampling.

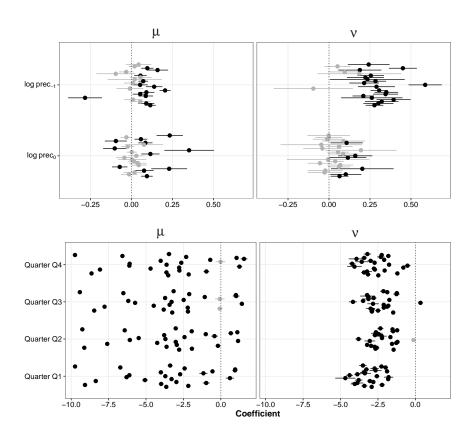


Figure S7: Graphical representation of coefficients from table S4. Top row: Effect of precipitation at day before sampling and at day of sampling. Bottom row: estimates for the four Quarters. Each dot represents one compound (in the order described in table S3). Coefficients where the CI encompasses zero are shown in gray colour. Coefficients are shown on the link scale (log for μ and logit for ν).

5 Pesticides in small streams

Table S5: Overview on RAC exceedances of the 76 compounds with more than 1000 measurements. No. = number of measurements; % RQ >1 = RAC exceedances; % RQ >1 | >LOQ= RAC exceedances as fraction of detects.

Name	No.	No.	%	No.	% RQ	% RQ
		>LOQ	>LOQ	RQ > 1	>1	>1
						>LOQ
2,4-D	12290	284	2.3	10	0.1	3.5
Aclonifen	9861	67	0.7	4	0.0	6.0
Azoxystrobin	7059	690	9.8	6	0.1	0.9
Benalaxyl	6964	10	0.1	0	0.0	0.0
$\operatorname{Bentazon}$	12429	2421	19.5	0	0.0	0.0
$\operatorname{Boscalid}$	9886	2296	23.2	0	0.0	0.0
Bromoxynil	9451	78	0.8	0	0.0	0.0
Carbendazim	3851	654	17.0	12	0.3	1.8
Chloridazon	15724	511	3.2	0	0.0	0.0
Chlorpyrifos	14704	954	6.5	954	6.5	100.0
Chlortoluron	18286	371	2.0	2	0.0	0.5
Clomazon	9268	440	4.7	0	0.0	0.0
Clopyralid	5520	107	1.9	0	0.0	0.0
Clothianidin	2409	154	6.4	123	5.1	79.9
Cypermetryn	1428	5	0.4	1	0.1	20.0
Cyprodinil	9779	118	1.2	0	0.0	0.0
Dicamba	7641	76	1.0	0	0.0	0.0
Difenoconazol	1644	11	0.7	2	0.1	18.2
Diflufenican	15457	1932	12.5	273	1.8	14.1
Dimefuron	7833	5	0.1	0	0.0	0.0
Dimethachlor	8858	344	3.9	0	0.0	0.0
Dimethenamid	9568	594	6.2	5	0.1	0.8
Dimethoat	14423	185	1.3	1	0.0	0.5
Dimethomorph	2316	91	3.9	0	0.0	0.0
Dimoxystrobin	3370	232	6.9	49	1.5	21.1
Diuron	18560	2336	12.6	40	0.2	1.7
Epoxiconazol	16454	621	3.8	7	0.0	1.1
Ethofumesat	20430	1078	5.3	0	0.0	0.0
Fenhexamid	2690	42	1.6	0	0.0	0.0

		400				
Fenpropimorph	12850	199	1.5	5	0.0	2.5
Fluazifop-P	4033	14	0.3	0	0.0	0.0
Fluazifop-P-butyl	1728	0	0.0	0	0.0	2.4
Fludioxonil	3203	42	1.3	1	0.0	2.4
Flufenacet	13509	798	5.9	1	0.0	0.1
Fluquinconazole	6762	117	1.7	0	0.0	0.0
Fluroxypyr	8096	378	4.7	0	0.0	0.0
Flurtamone	16958	638	3.8	2	0.0	0.3
Flusilazol	5257	53	1.0	1	0.0	1.9
Glyphosate	3557	1455	40.9	1	0.0	0.1
Imidacloprid	3169	192	6.1	169	5.3	88.0
Ioxynil	8114	20	0.2	0	0.0	0.0
Isoproturon	19112	4164	21.8	92	0.5	2.2
Kresoxim-methyl	6929	14	0.2	0	0.0	0.0
Lenacil	13837	183	1.3	0	0.0	0.0
MCPA	12773	1687	13.2	2	0.0	0.1
Mecoprop	12521	1552	12.4	0	0.0	0.0
Metalaxyl	14460	299	2.1	0	0.0	0.0
Metamitron	15390	613	4.0	0	0.0	0.0
Metazachlor	21906	2015	9.2	55	0.3	2.7
Methamidophos	1303	0	0.0	0	0.0	
Methobromuron	14968	24	0.2	1	0.0	4.2
Metribuzin	15411	192	1.2	15	0.1	7.8
Napropamid	9914	269	2.7	1	0.0	0.4
Nicosulfuron	5172	288	5.6	77	1.5	26.7
Penconazol	4846	159	3.3	0	0.0	0.0
Pendimethalin	16997	328	1.9	4	0.0	1.2
Pethoxamid	3102	37	1.2	0	0.0	0.0
Phoxim	1492	0	0.0	0	0.0	
Picolinafen	8901	11	0.1	2	0.0	18.2
Picoxystrobin	3620	7	0.2	0	0.0	0.0
Pirimicarb	11330	232	2.0	27	0.2	11.6
Prochloraz	5795	33	0.6	0	0.0	0.0
Propiconazol	14250	818	5.7	7	0.0	0.9
Propyzamid	11937	453	3.8	0	0.0	0.0
Prosulfocarb	5001	126	2.5	0	0.0	0.0
Pyrimethanil	8136	122	1.5	0	0.0	0.0
Quinmerac	7291	989	13.6	0	0.0	0.0
Rimsulfuron	1240	2	0.2	0	0.0	0.0
Spiroxamin	2469	109	4.4	1	0.0	0.9
Tebuconazol	16584	1024	6.2	26	0.2	2.5
Terbuthylazin	22568	3370	14.9	35	0.2	1.0
Thiacloprid	3540	85	2.4	85	2.4	100.0
Thiamethoxam	1853	39	2.1	7	0.4	17.9

Triadimenol	3067	51	1.7	0	0.0	0.0
Triazophos	3588	2	0.1	1	0.0	50.0
Trifloxystrobin	3674	10	0.3	1	0.0	10.0

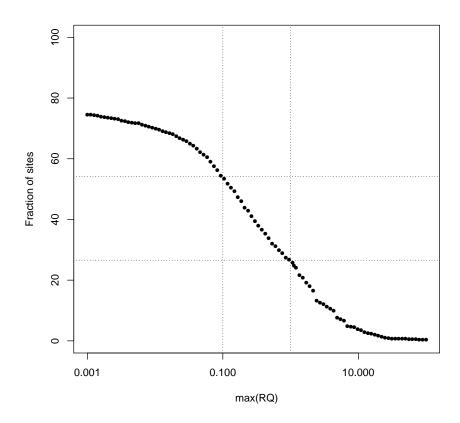


Figure S8: Cumulative distribution of sites exceeding RAC. Dotted lines indicate fraction of sites exceeding a RQ of 1 and 0.1. 23% of the sites showed no detection of compounds with available RAC values and are not shown due to logarithmic x-axis.

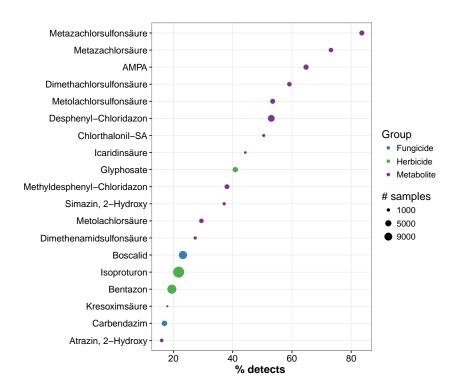


Figure S9: Proportion of samples with detects in small streams. Only Compounds with more than 100 samples and 15% of detects are shown.

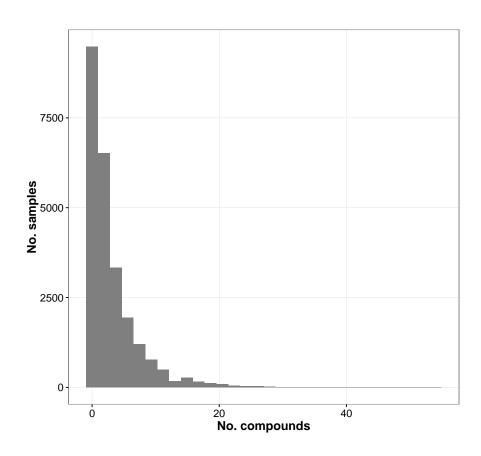


Figure S10: Distribution of the number of quantified compounds in the samples.