Modelling biocide and herbicide concentrations in catchments of the Rhine basin

Supplementary material

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Content:

Part I: Model description

Appendix A1: CrossWater model framework

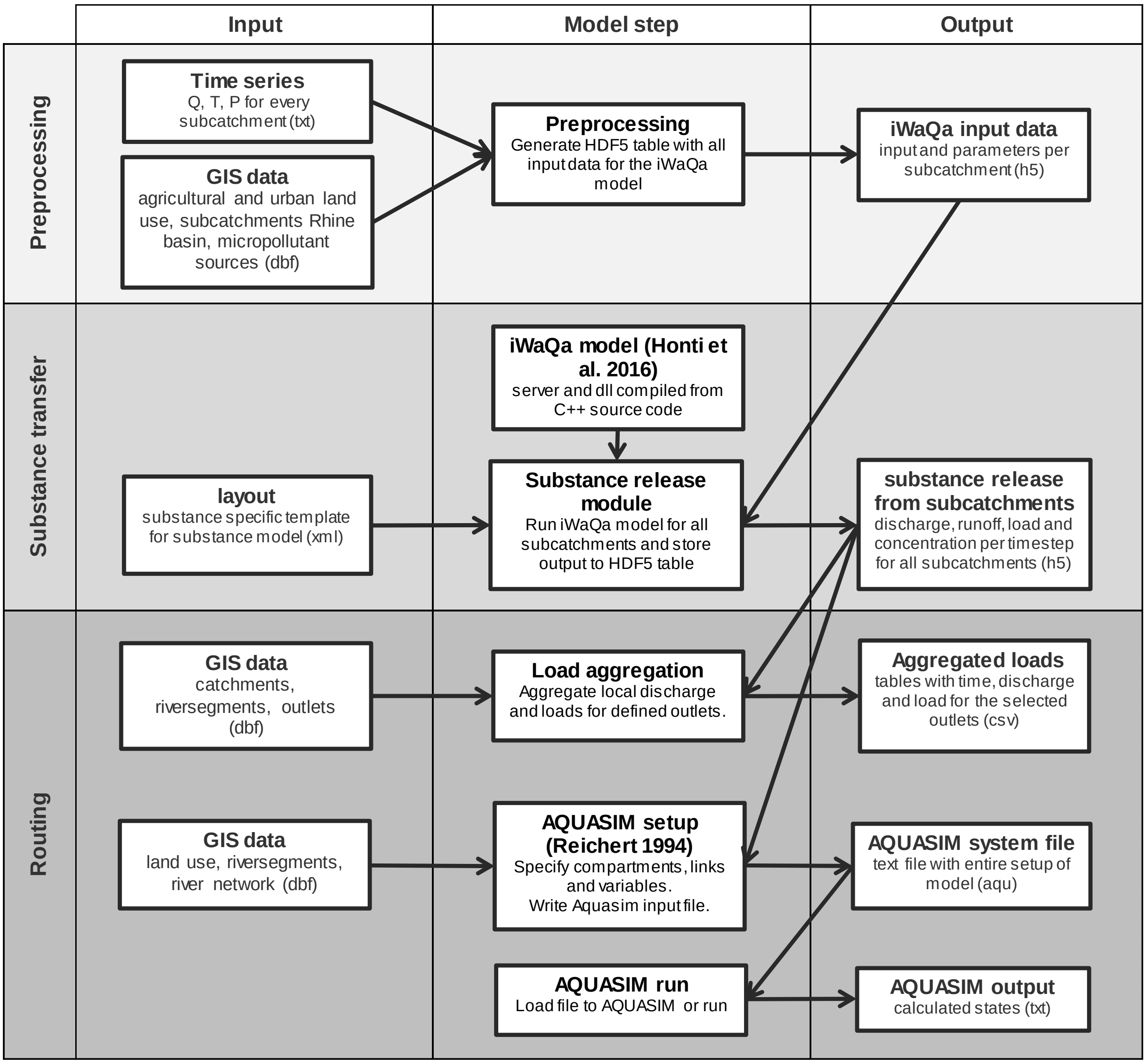


Figure S1 : Flow chart of CrossWater framework to model herbicide and biocide concentrations in the Rhine basin. (txt): text file, (csv): CSV-formatted text file, (dbf): dBASE database file, (h5): Hierarchical Data Format HDF5, (aqu): AQUASIM job file

Appendix A2: Conceptual model of the spatial representation

Due to the usage of CCM2 subcatchments (Vogt et al., 2007) and the aggregation procedure to guarantee a reasonable minimal subcatchment area as described in the main text, not all joints of the hydrologic tree were preserved. To tackle this, we conceptually differentiate between catchments that don't have any lateral inputs (e.g. headwaters or higher order streams sections without tributaries) and the inter-catchment areas (see Figure S2). Mass transfer from both classes of subcatchments is simulated the same way. For simulating herbicide losses however, there is a slight difference in that for catchments the driving force is the (observed) discharge at the catchment outlet (see below), while for inter-catchment areas lateral discharge has to be determined in another way (as outlined below).

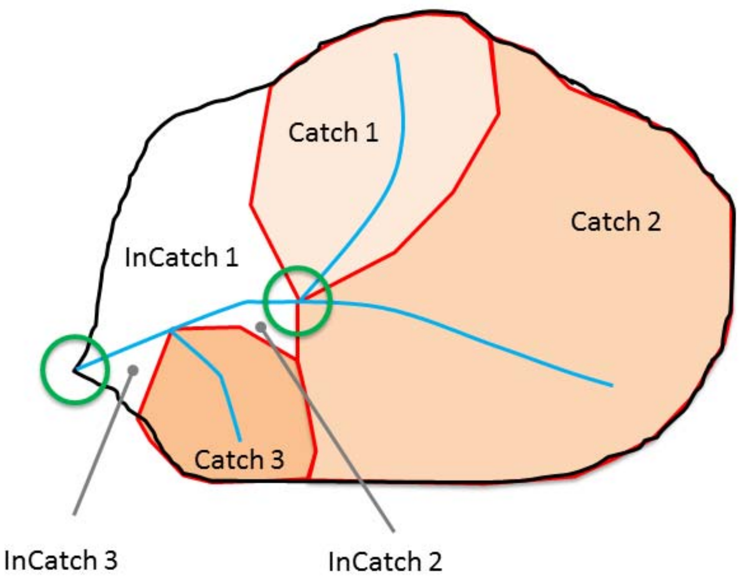
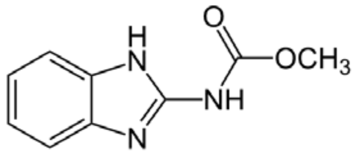
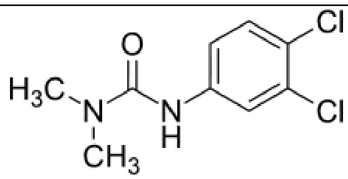
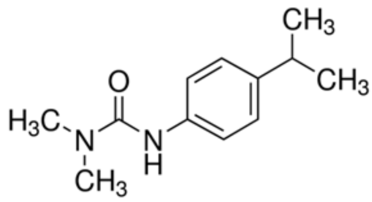
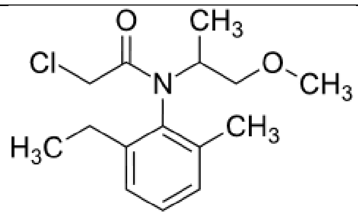
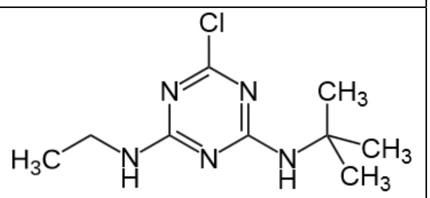


Figure S2: Schematic representation of the spatial units covering the basin. The example basin consists of the three catchment areas Catch 1 to Catch 3 and three inter-catchment areas.

Appendix A3: Model compounds

Table S1 : Characterisation of the model compounds. Sources: http://gestis.itrust.de (last access: 8. Aug. 2017).

Compound	Use	CAS No	Log K_{ow}	Structure
Carbendazim	Biocide, (herbicide)	10605-21-7	1.43	
Diuron	Biocide, (herbicide)	330-54-1	2.78	
Isoproturon	Herbicide	34123-59-6	2.87	
S-Metolachlor	Herbicide	87392-12-9	3.13	
Terbuthylazine	Herbicide	5915-41-3	3.06	

Appendix A4: Data sources of discharge, precipitation and temperature

Table S2 : Discharge, precipitation and temperature data sources for the Rhine basin

	Germany	**Switzerland**	**Other**
Discharge			
	Bundesamt für Gewässerkunde Landesamt für Umwelt- und Arbeitsschutz Saarland Bayerisches Landesamt für Umwelt Landesamt für Natur, Umwelt und Verbraucherschutz Nordrhein-Westfalen Hessisches Landesamt für Umwelt und Geologie Landesanstalt für Umwelt, Messungen und Naturschutz Baden Württemberg Landesamt für Umwelt, Wasserwirtschaft und Gewerbeaufsicht Rheinland-Pfalz Thüringer Landesanstalt für Umwelt und Geologie	Bundesamt für Umwelt Departement Bau, Verkehr und Umwelt Aargau Amt für Wasser und Abfall des Kantons Bern Amt für Umweltschutz und Energie Basel-Land Amt für Natur und Umwelt Graubünden Umwelt und Energie Luzern Département du développement territorial et de l'environnement Neuchâtel Amt für Umwelt St. Gallen Amt für Umwelt Solothurn Amt für Umwelt Thurgau Amt für Abfall, Wasser Energie und Luft Zürich Département du territoire et de l'environnement Waadt	Austria: Amt der Vorarlberger Landesregierung France: Ministry of Ecology, Sustainable Development and Energy Luxembourg: Administration de la Gestion de l'Eau
Precipitation			
	Deutscher Wetterdienst, RADOLAN Deutscher Wetterdienst, Stationen	MeteoSwiss, CombiPrecip	Luxembourg: Atlas hydro-climatologique du Grand-Duché de Luxembourg 2010
Temperature			
		MeteoSwiss, TabsD	EU: European Climate Assessment & Dataset, E-OBS

Appendix A5:

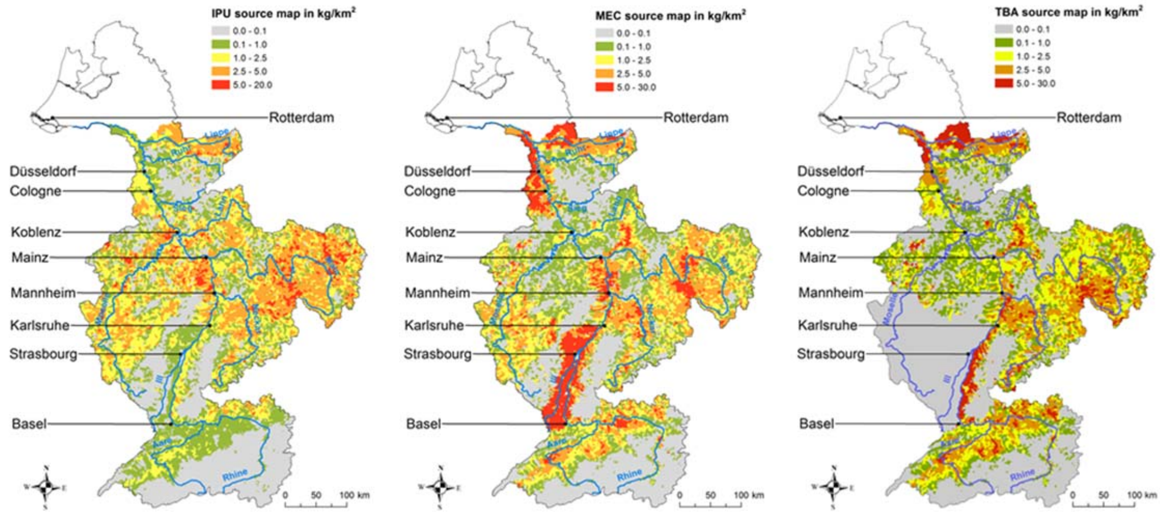


Figure S3: Spatial distribution of the herbicide applications across the Rhine basin. IPU: isoproturon, MEC: S-metolachlor, TBA: terbuthylazine. (Base data: (Eurostat, 2011;European Environment Agency EEA, 2012;Swisstopo, 2007;Swiss Federal Office for the Environment (FOEN), 2012;Swiss Federal Statistical Office FSO, 2012, 2011;Vogt et al., 2007)

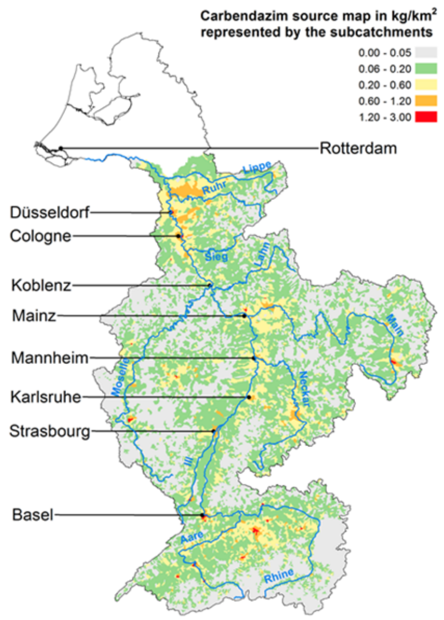


Figure S4: Spatial distribution of the carbendazim applications across the Rhine basin. The spatial pattern for diuron is the same because of the absence of spatially distributed input data. (Base data: (Swiss Federal Office for the Environment (FOEN), 2012;Herlyn and Maurer, 2007;European Environment Agency EEA, 2012;Eurostat, 2011;Vogt et al., 2007;Swisstopo, 2007).

Appendix A6: Statistical relationships between footprint of buildings, population density and area of facades in Switzerland.

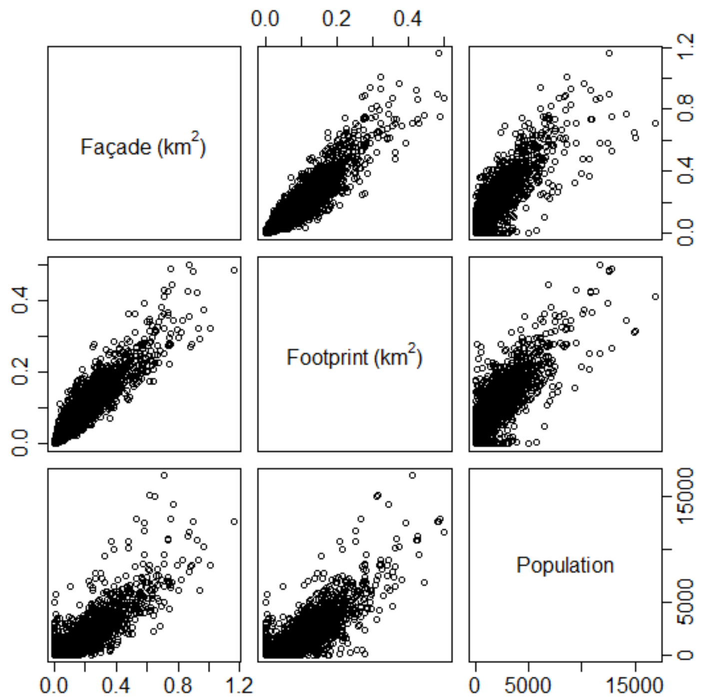


Figure S5: Relations between façade, footprint and population in the subcatchments of Switzerland

Appendix A7: Calibration, validation and prediction sites

Table S3 : Characterization of calibration, validation and prediction catchments

	Catchment/ River	Abbr.	Reference	IPU	TBA	CBZ	DIU	MCP	Year	Area [km^2]	agricultural land use [km^2]	housing [km^2]	population
Calibration													
	Ossingen	oss	Doppler et al. 2012	x	x				2009	1.2	1.1	-	-
	Summerau	sum	Freitas et al. 2008						2003	0.5	0.04	-	-
	Moenchaltdorf	moe	Wittmer et al. 2010	x		x	x	x	2007	24.6	4.7	0.5	12'000
Validation													
	Furtbach	fch	NAWA SPEZ	x	x	x	x	x	2012	31	14	1.6	31'570
	Limpach	lch	NAWA SPEZ	x	x	x	x	x	2012	74	43	1	7'560
	Mentue	mnt	NAWA SPEZ	x	x	x	x	x	2012	100	42	1	9'300
	Salmsacher Aach	smr	NAWA SPEZ	x	x	x	x	x	2012	54	33	1.7	17'326
	Surb	srb	NAWA SPEZ	x	x	x	x	x	2012	68	36	1.4	22'780
	Thur	thr	NADUF	x		x	x	x	2009	1'735	873	33	403'028
	Toess	tss	NADUF	x		x	x	x	2009	432	175	11	197'032
	Glatt	glt	NADUF	x		x	x	x	2009	413	183	20	405'702
	Murg	mrg	NADUF	x		x	x	x	2009	212	118	5.3	68'145
	Sitter	str	NADUF	x		x	x	x	2009	96	21	0.9	9'704
	Rhine-Reckingen	rhn-rkg	NADUF	x		x	x	x	2009	14'721	5'261	175	2'946'907
	Rhine-Basel	imrs	IMRS	x	x	x	x	x	2011	35'899	12'009	503	7'786'398
Validation/Prediction													
	Aare	are		(x)	(x)				2011	17'623	5'399	270	3'957'080
	Neckar	nkr		(x)	(x)				2011	13'897	5'536	318	5'787'797
	Main	man		(x)	(x)				2011	27'235	11'450	466	7'194'708
	Moselle	msl		(x)	(x)				2011	28'199	12'152	347	4'646'691
	Rhine-Lobith	rhn-lbt	ICPR	x	(x)				2011	160'221	64'574	3028	50'210'696

x modelled and validated

(x) only prediction

Appendix A8: Calibration of the herbicide substance transfer module

Table S4 : Prior distributions for the calibration of the herbicide substance transfer module

parameter	distribution	unit		isoproturon	metolachlor	terbuthylazine
ρ	uniform	-	mean	0.068	0.422	0.200
			max	1	1	1
			min	0	0	0
k_d, k_{error}	lognormal	1/d	mean	0.00924	0.0077016	0.0315
			stdv	0.01	0.01	0.01
k_{w-s}	uniform	1/d	mean	0.027	0.03	0.05
			max	1	1	1
			min	1.00E-20	1.00E-20	1.00E-20
k_{s-w}	uniform	1/d	mean	0.00038	0.002	8.00E-04
			max	1	1	1
			min	1.00E-20	1.00E-20	1.00E-20
T_{obj}	normal	°C	mean	200	750	750
			stdv	100	100	100
C_{back}	lognormal	ng/l	mean	7	7	5
			stdv	2	2	2
ε	normal	s/m2	mean	2.30E+07	3.17E+07	1.29E+09
			stdv	2.16E+07	2.45E+07	7.18E+08
ρ_{error}	normal		mean	300	300	300
			stdv	50	50	50

Table S5: Prior distributions for the biocide substance transfer module.

parameter	distribution	unit		diuron	carbendazim
β	uniform	mm/d	mean	1.18E-04	7.35E-05
			max	3.48E-04	2.37E-04
			min	1.58E-05	7.90E-06

Appendix A9: River segments for the Aquasim application

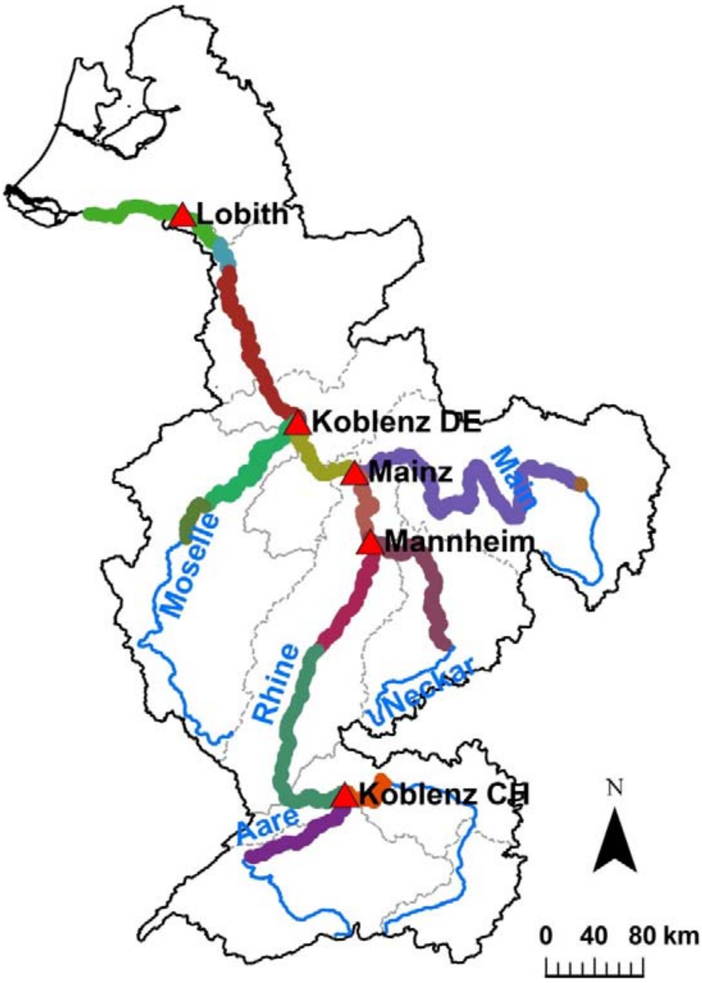


Figure S6 : Rhine basin with subdivisions into the major catchments. The Rhine and the main tributaries are divided into 15 compartments (colored river segments) for the hydraulic routing with AQUASIM. Concentration time series are investigated at the displayed cities along river system. (Base data: GEOSTAT 2011; JRC 2007, 2008; Swisstopo 2007).

Part III: Results

Appendix A10: Parameter estimates

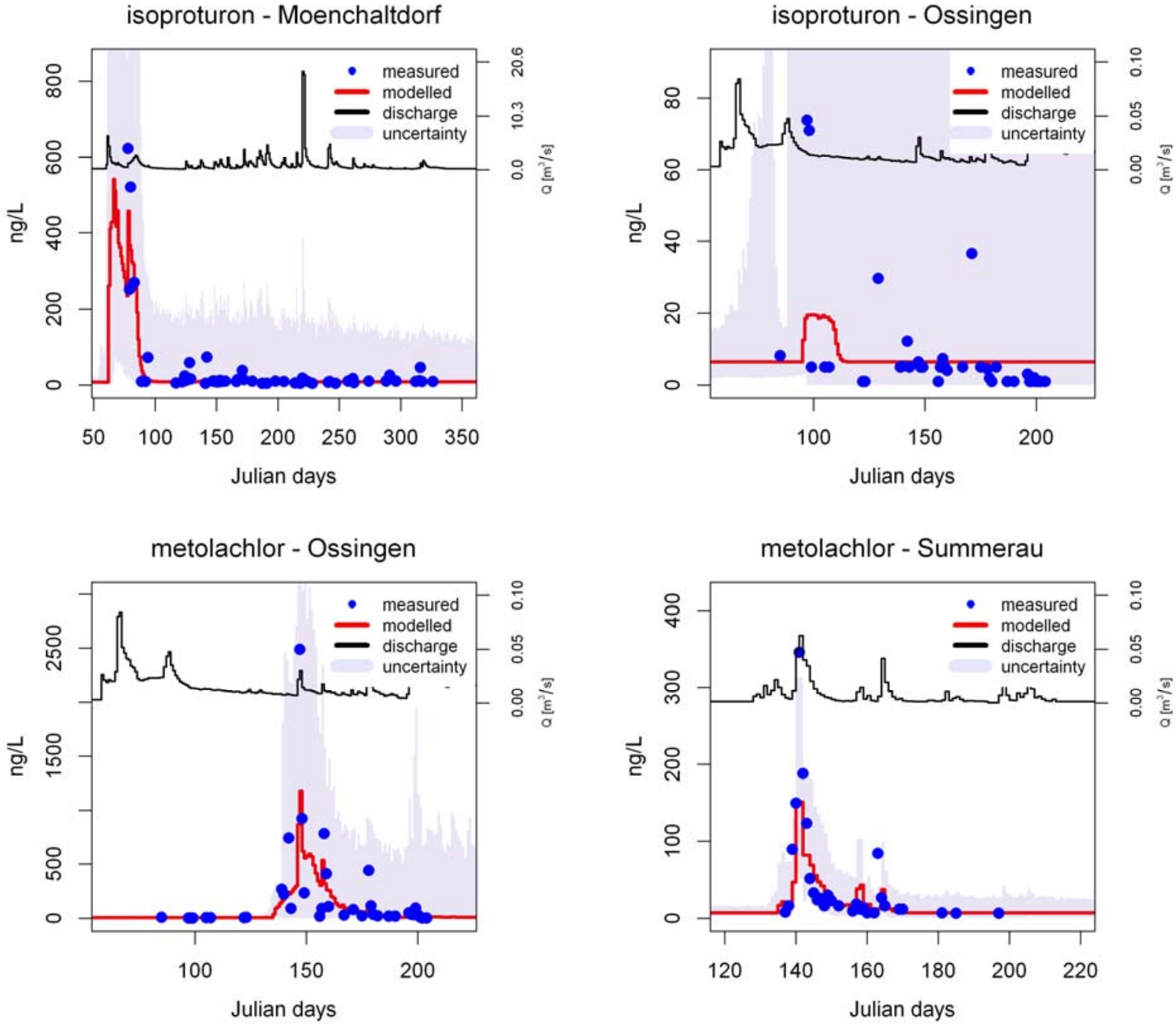
Table S6 : Calibrated parameters for the herbicide substance transfer module. Abbreviations for study catchments: moe: Mönchaltdorf, oss: Ossingen, sum: Summerau.

parameter	unit	isoproturon		metolachlor		terbuthylazine	
		moe	oss	oss	summ	moe	oss
ρ	-	0.45	0.08	0.52	0.86	0.95	0.02
k_d	1/d	6.6E-03	6.0E-03	4.7E-03	4.8E-03	3.0E-02	3.0E-02
k_{w-s}	1/d	0.22	0.94	0.10	0.20	0.08	0.07
k_{s-w}	1/d	9.4E-05	2.5E-06	5.5E-05	2.3E-05	4.8E-04	2.6E-05
T_{obj}	°C	154	252*	812*	576*	737	812*
C_{back}	ng/l	8.1	6.4	6.7	7.0	4.6	4.7
ε	s/m^2	2.6E+07	2.3E+07	2.9E+07	8.6E+06	8.9E+06	9.3E+08
k_{error}	1/d	8.8E-04	3.5E-03	3.4E-03	2.0E-03	6.8E-03	1.3E-02
ρ_{error}	-	357	311	315	412	458	376
σ_{error}	-	1.31	2.45	0.78	1.17	575.87	14.21

Table S7 : Calibrated parameters for the biocide substance transfer module.

parameter	unit	carbendazim	diuron
		moe	moe
β	mm/d	5.6E-05	8.9E-05
σ_{error}	-	4.8	5.5

Appendix A11: Time-series of observed and calibrated concentrations



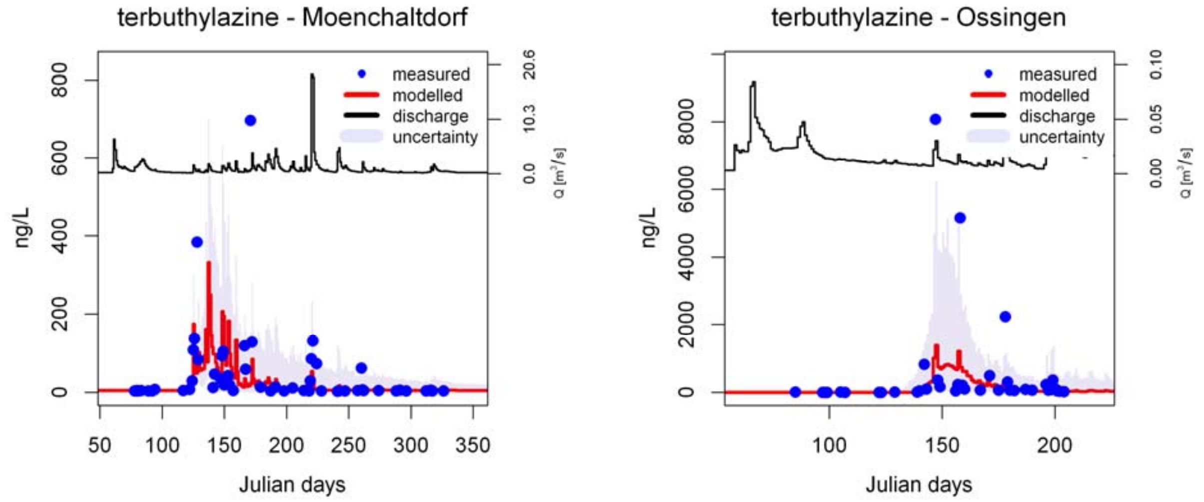


Figure S7 : Measured and modelled herbicide concentrations for the calibration sites. Note that the axis differ for the sites.

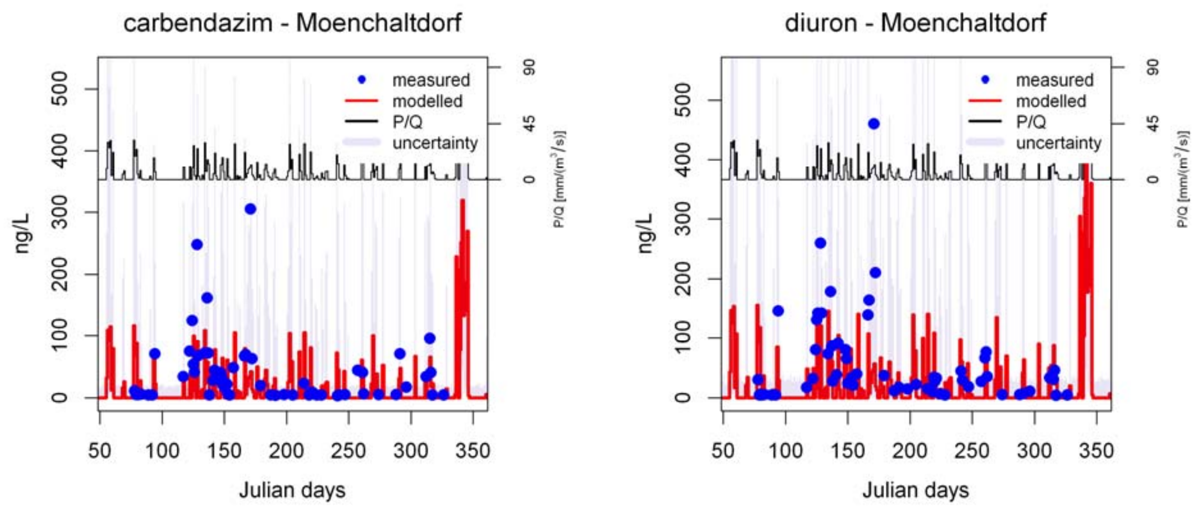


Figure S8 : Measured and modelled biocide concentrations for the calibration sites.

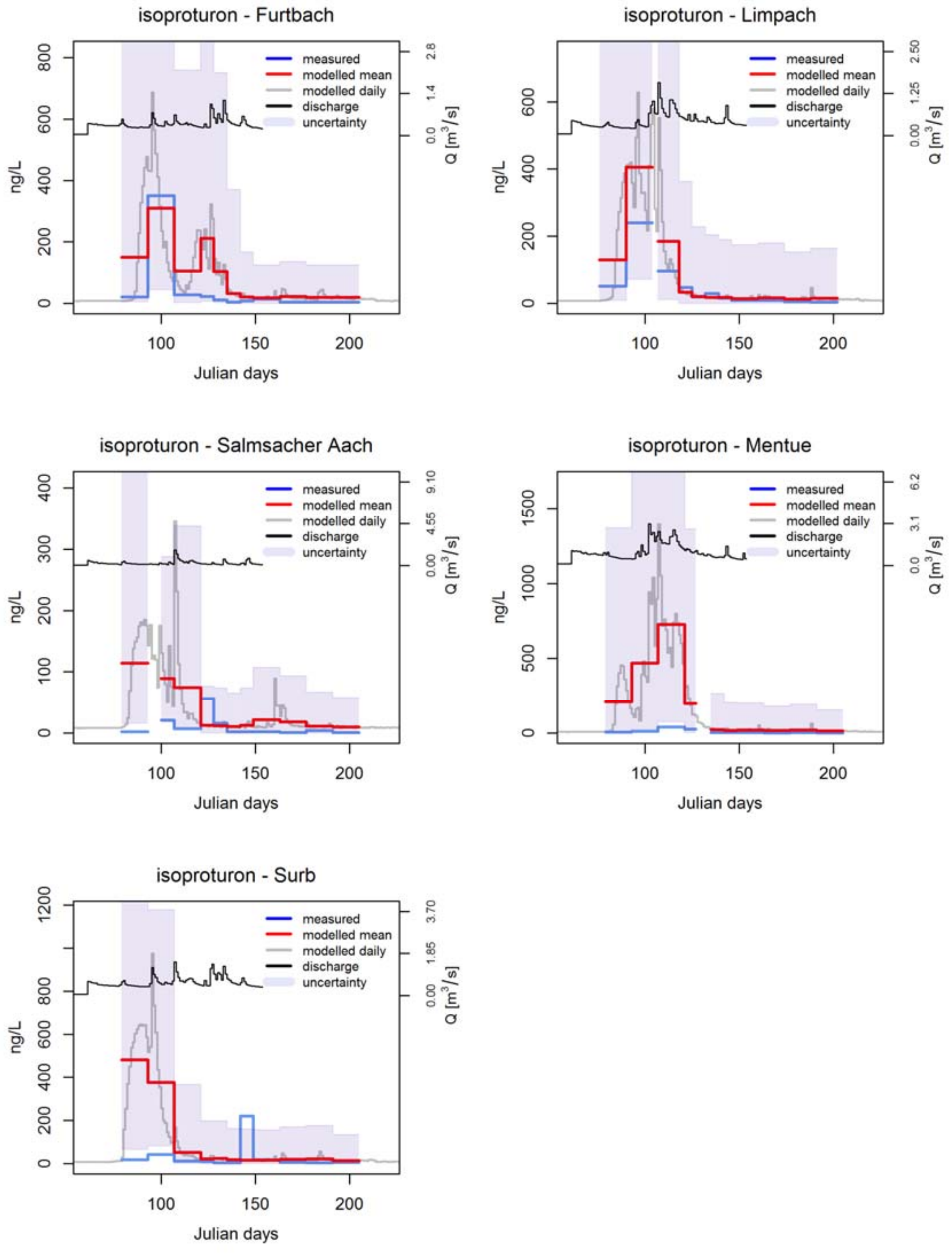


Figure S9 : Validation of the isoproturon predictions with the parameter set calibrated at Mönchaltdorf. Note that the axis differ for the sites.

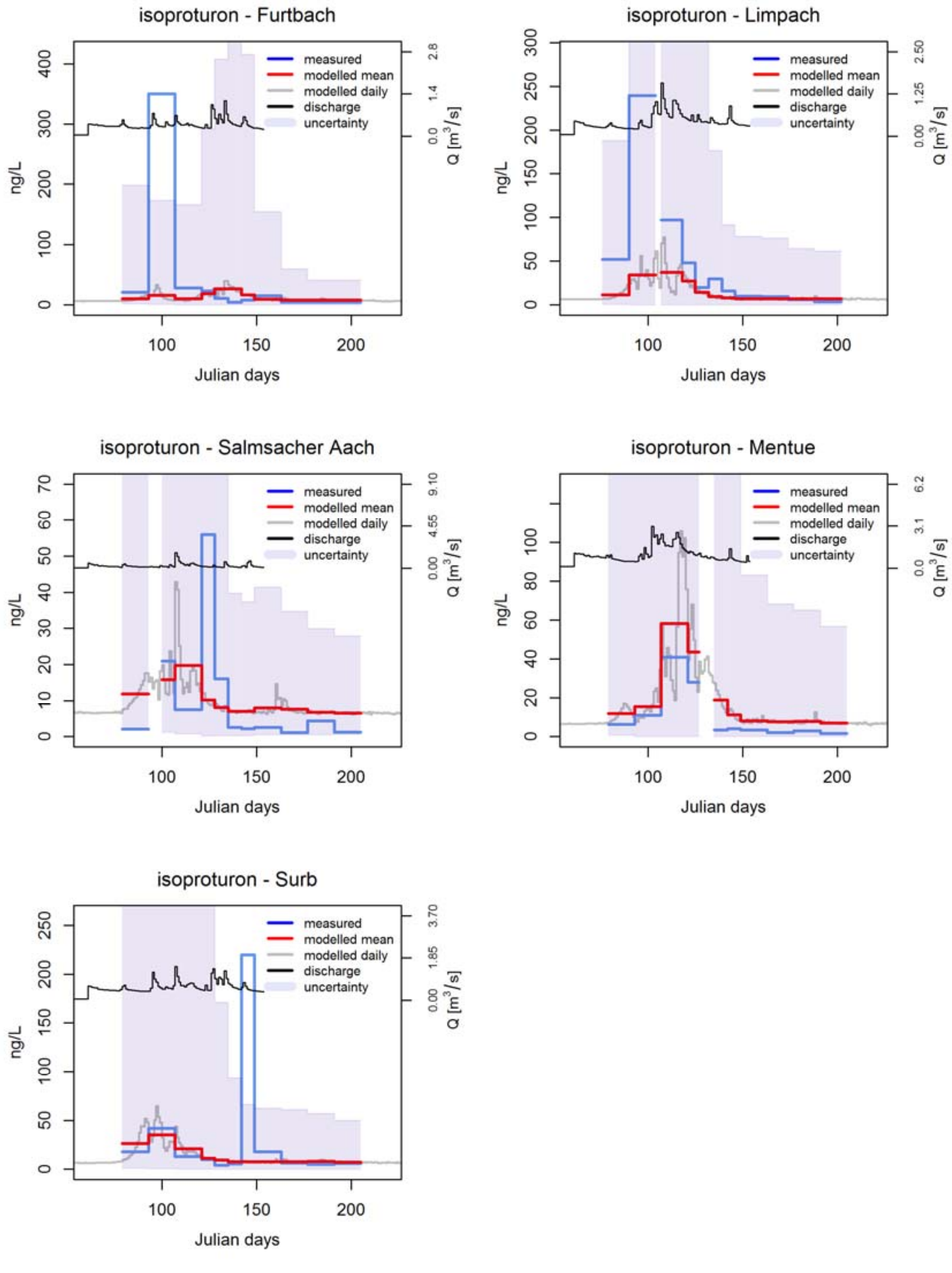


Figure **S10** : Validation of the isoproturon predictions with the parameter set calibrated at Ossingen. Note that the axis differ for the sites.

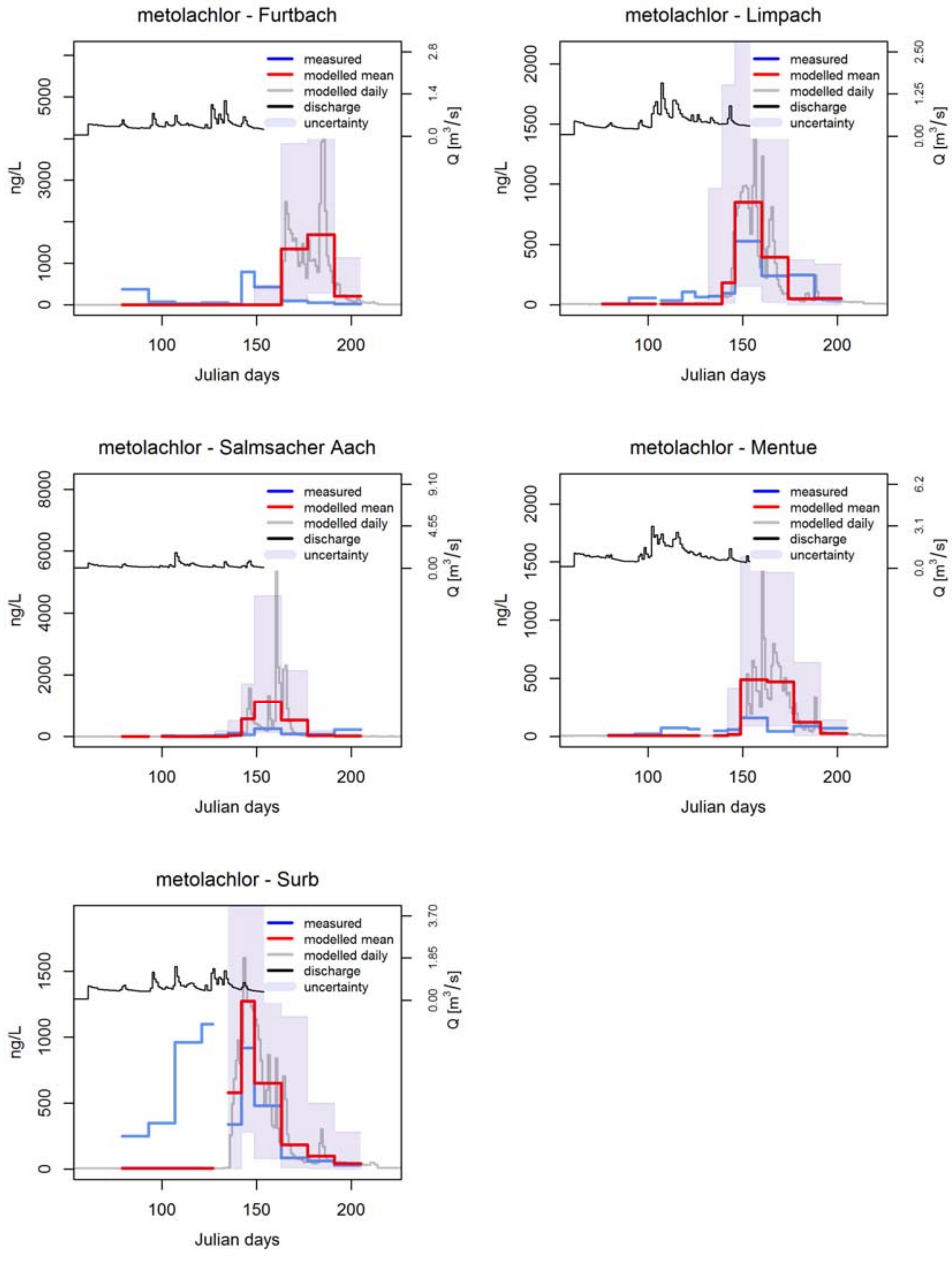


Figure S11 : Validation of the metolachlor predictions with the parameter set calibrated at Ossingen. Note that the axis differ for the sites.

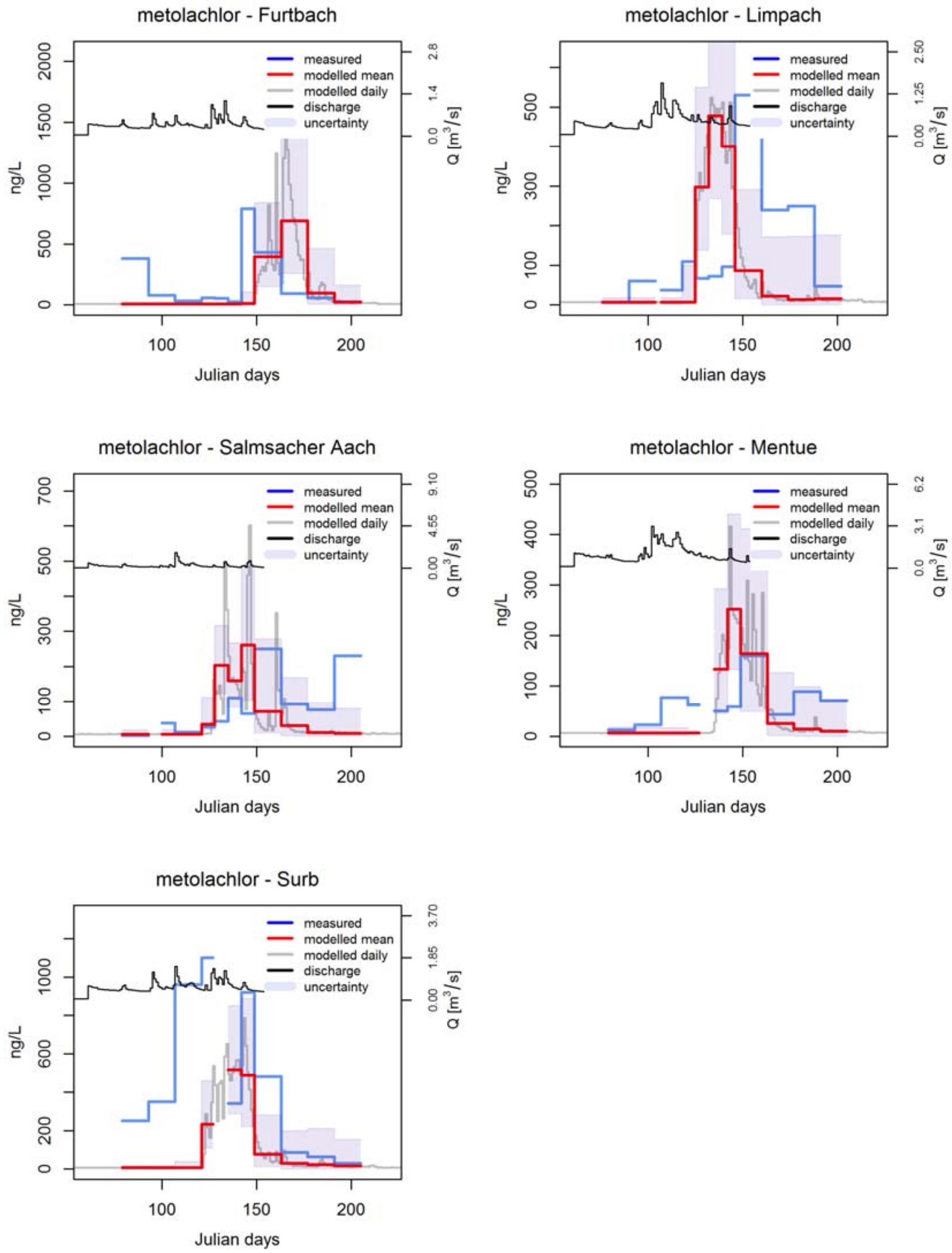


Figure S12 : Validation of the metolachlor predictions with the parameter set calibrated at Summerau. Note that the axis differ for the sites.

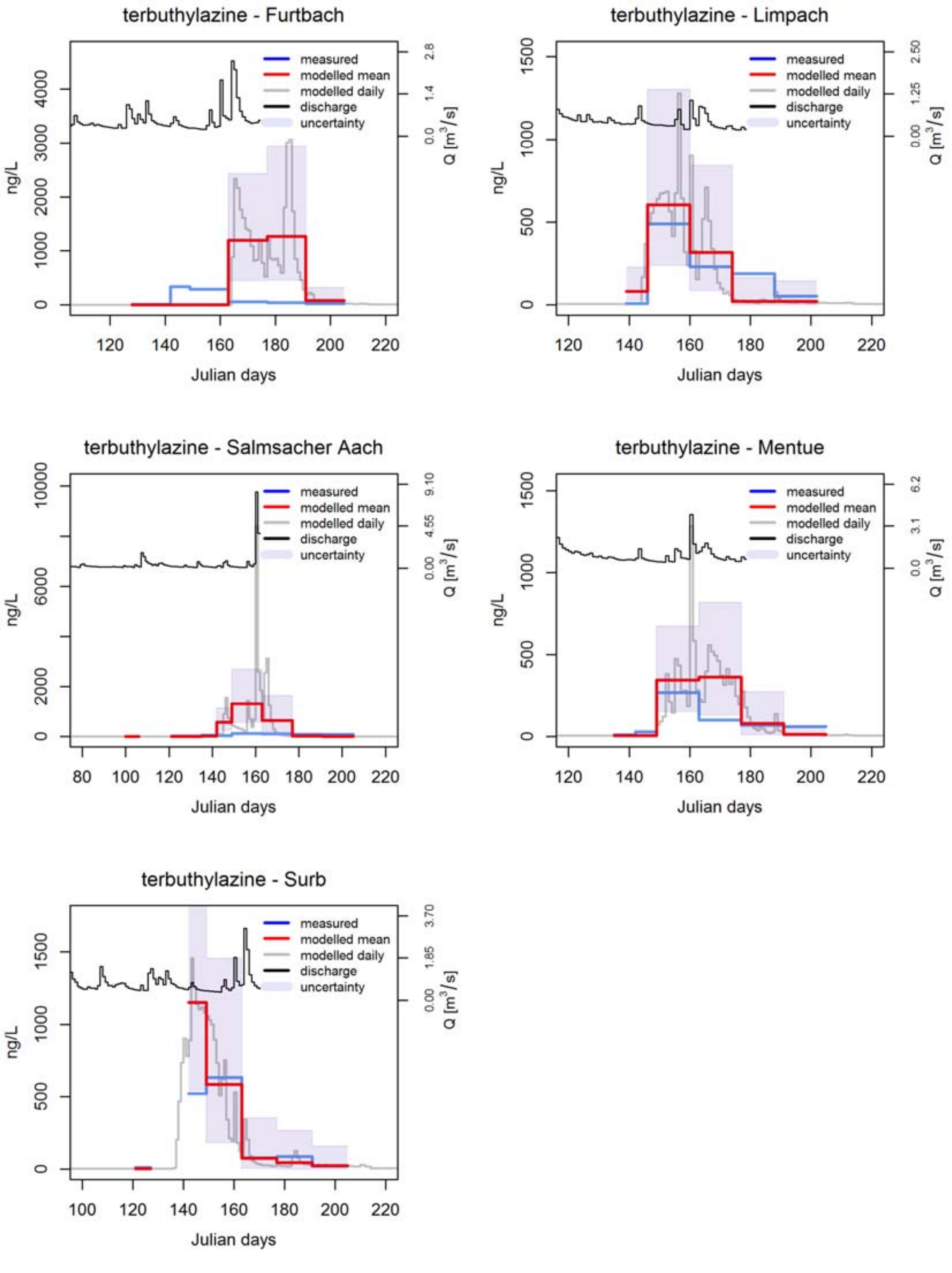


Figure S13 : Validation of the terbuthylazine predictions with the parameter set calibrated at Mönchaltdorf. Note that the axis differ for the sites.

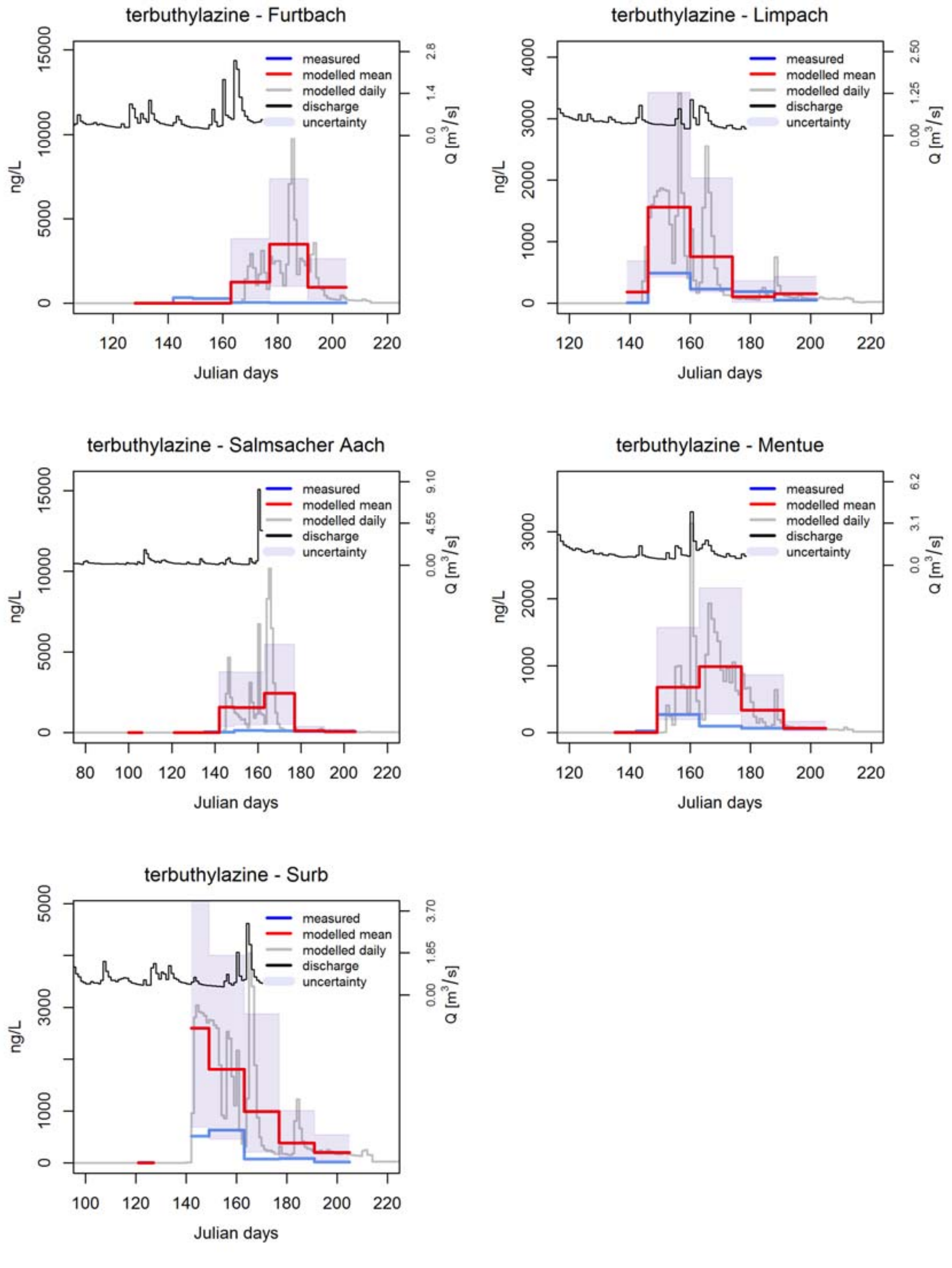


Figure S14 : Validation of the terbuthylazine predictions with the parameter set calibrated at Ossingen. Note that the axis differ for the sites.

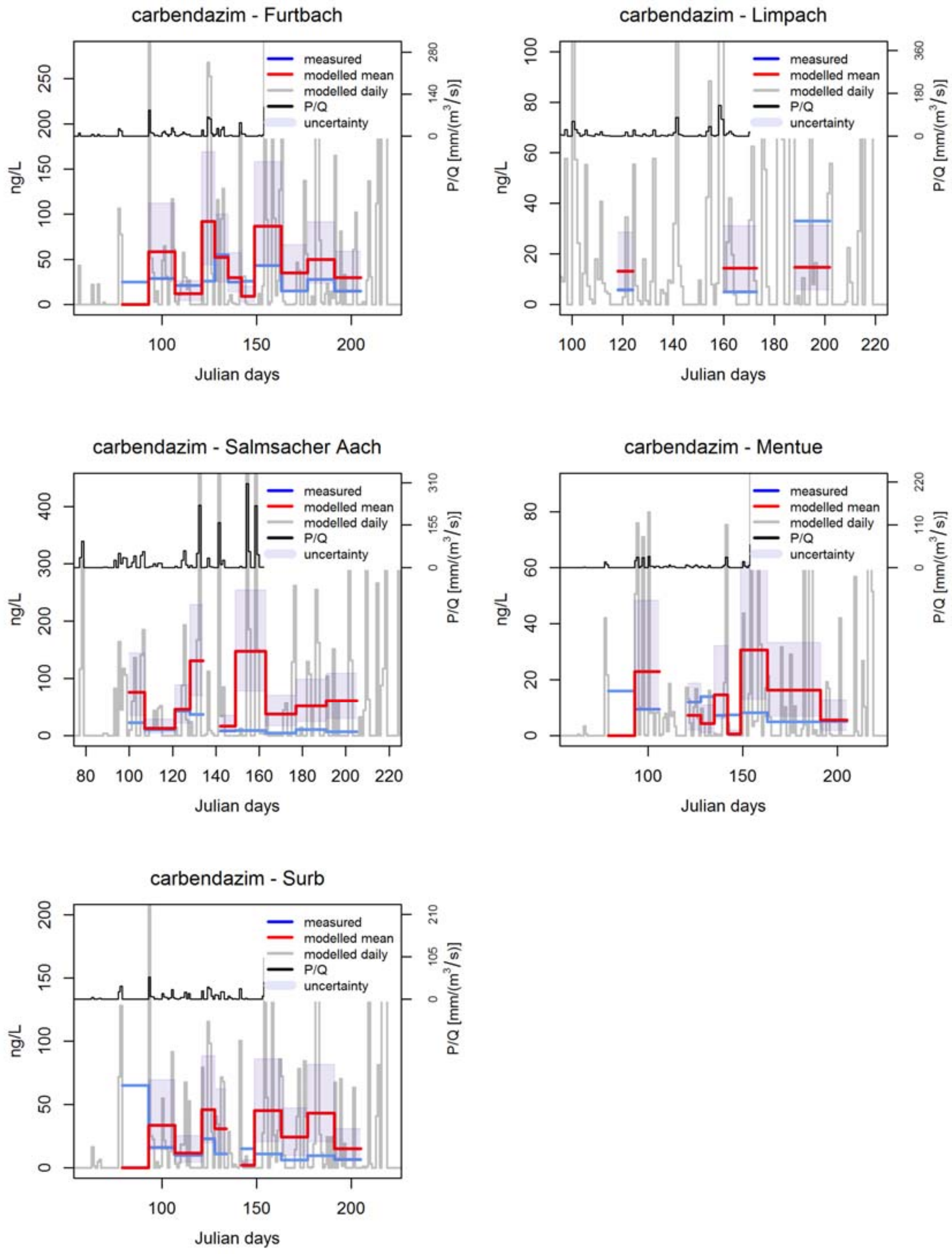


Figure S15 : Validation of the carbendazim predictions with the parameter set calibrated at Mönchaltdorf. Note that the axis differ for the sites.

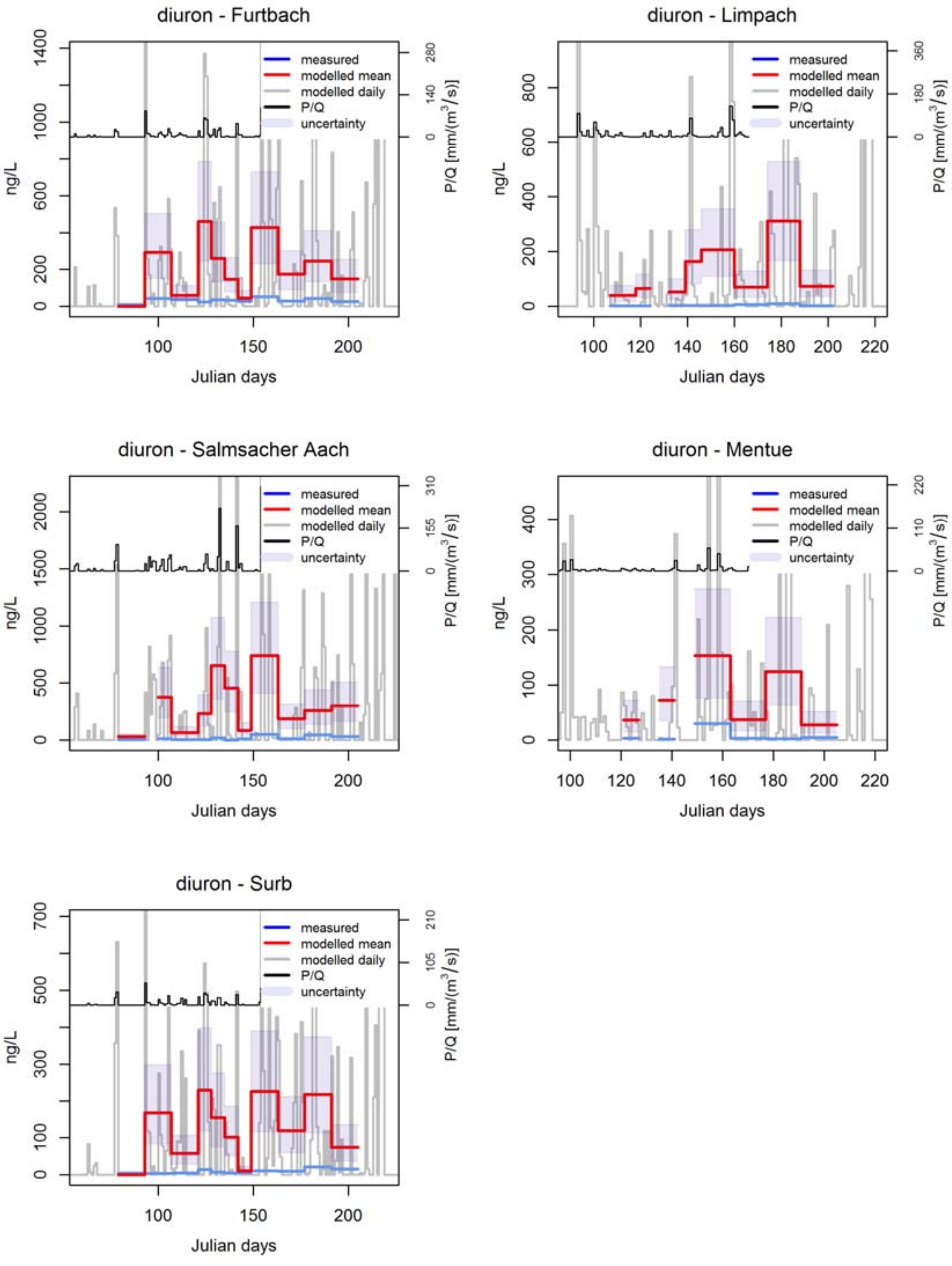


Figure S16 : Validation of the diuron predictions with the parameter set calibrated at Mönchaltdorf. Note that the axis differ for the sites.

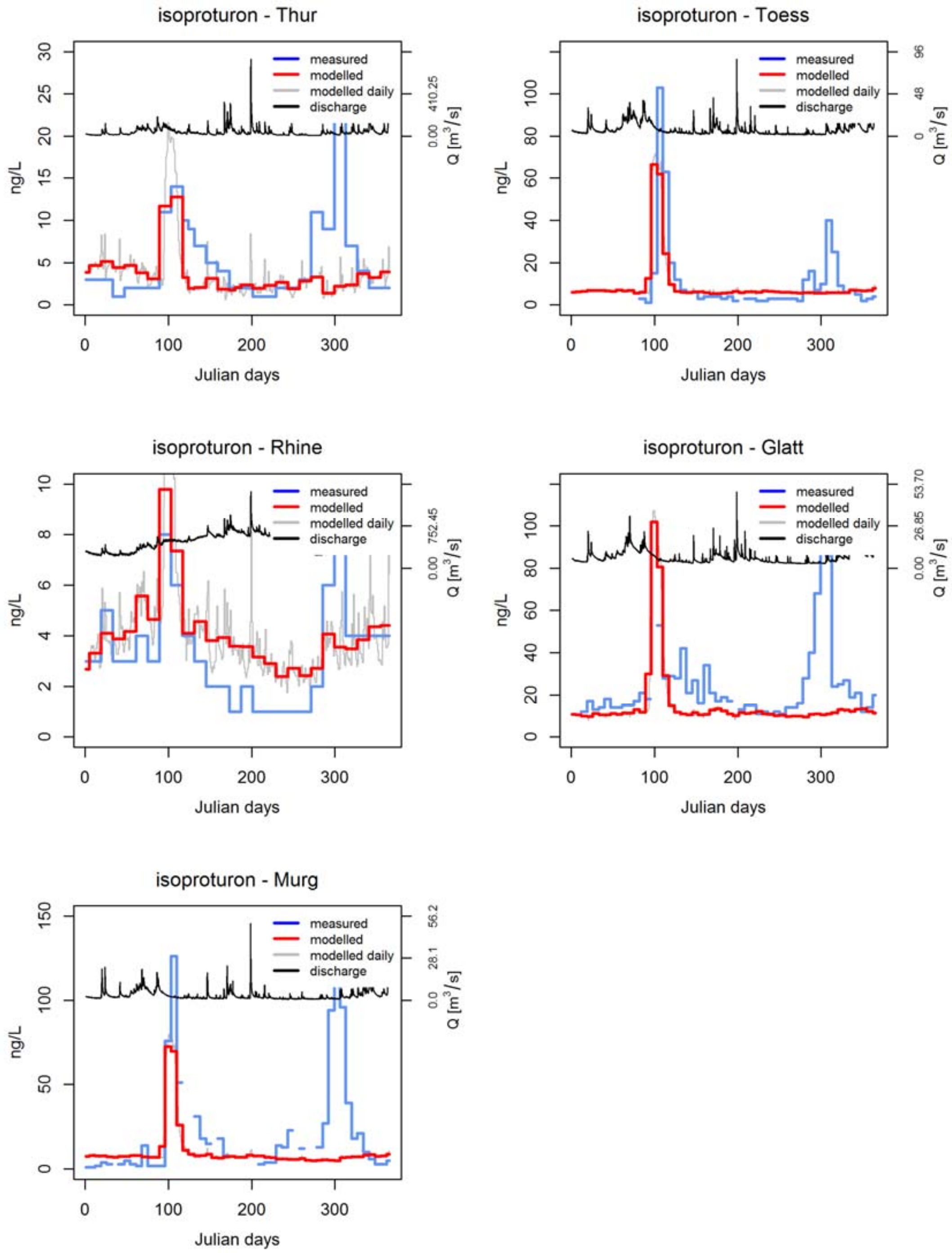


Figure S17 : Validation of isoproturon at the NADUF catchments with the calibrated parameter set from Mönchaltdorf. Note that the axis differ for the sites.

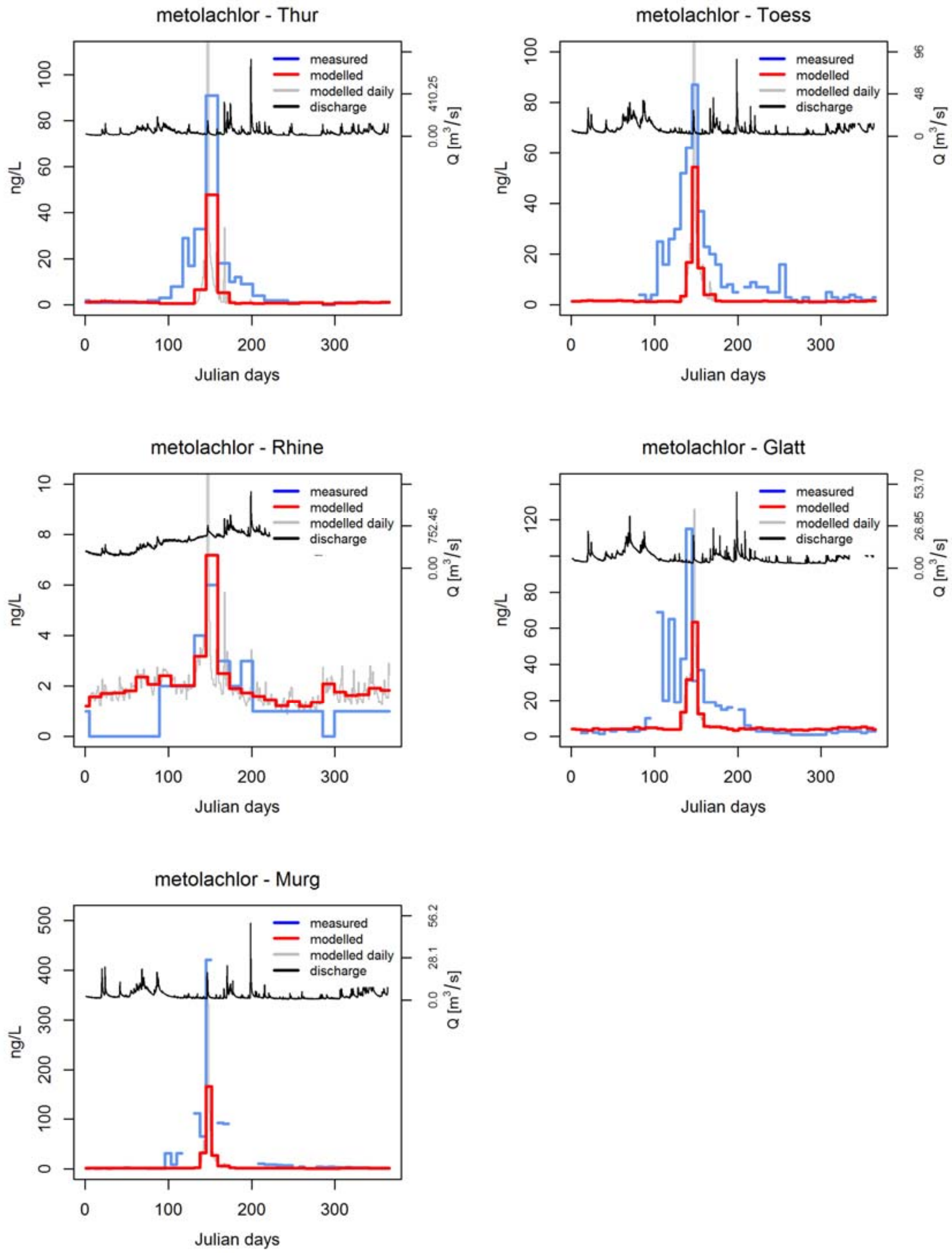


Figure S18 : Validation of metolachlor at the NADUF catchments with the calibrated parameter set from Summerau. Note that the axis differ for the sites.

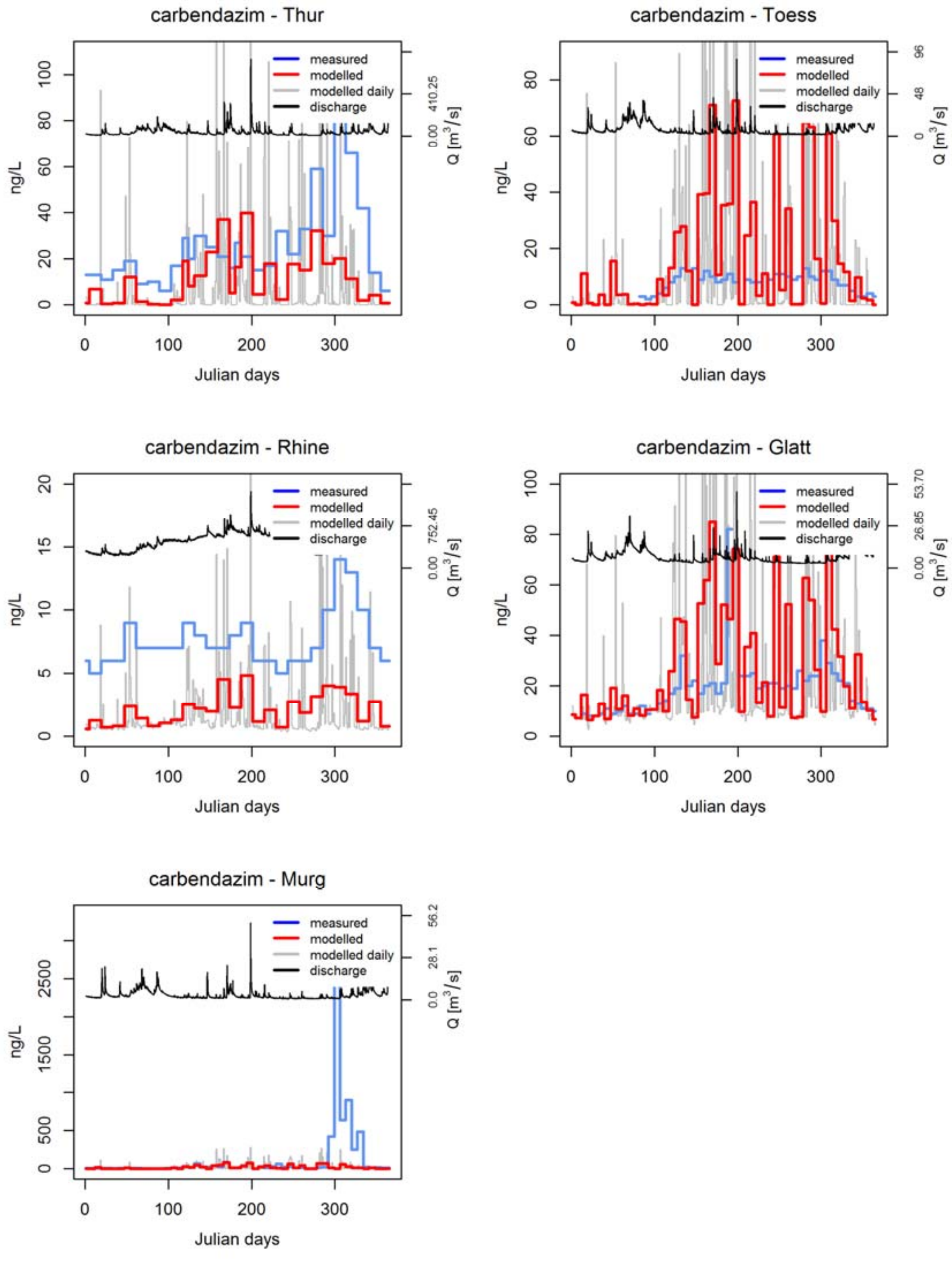


Figure S19 : Validation of carbendazim at the NADUF catchments with the calibrated parameter set from Mönchaltdorf. Note that the axis differ for the sites.

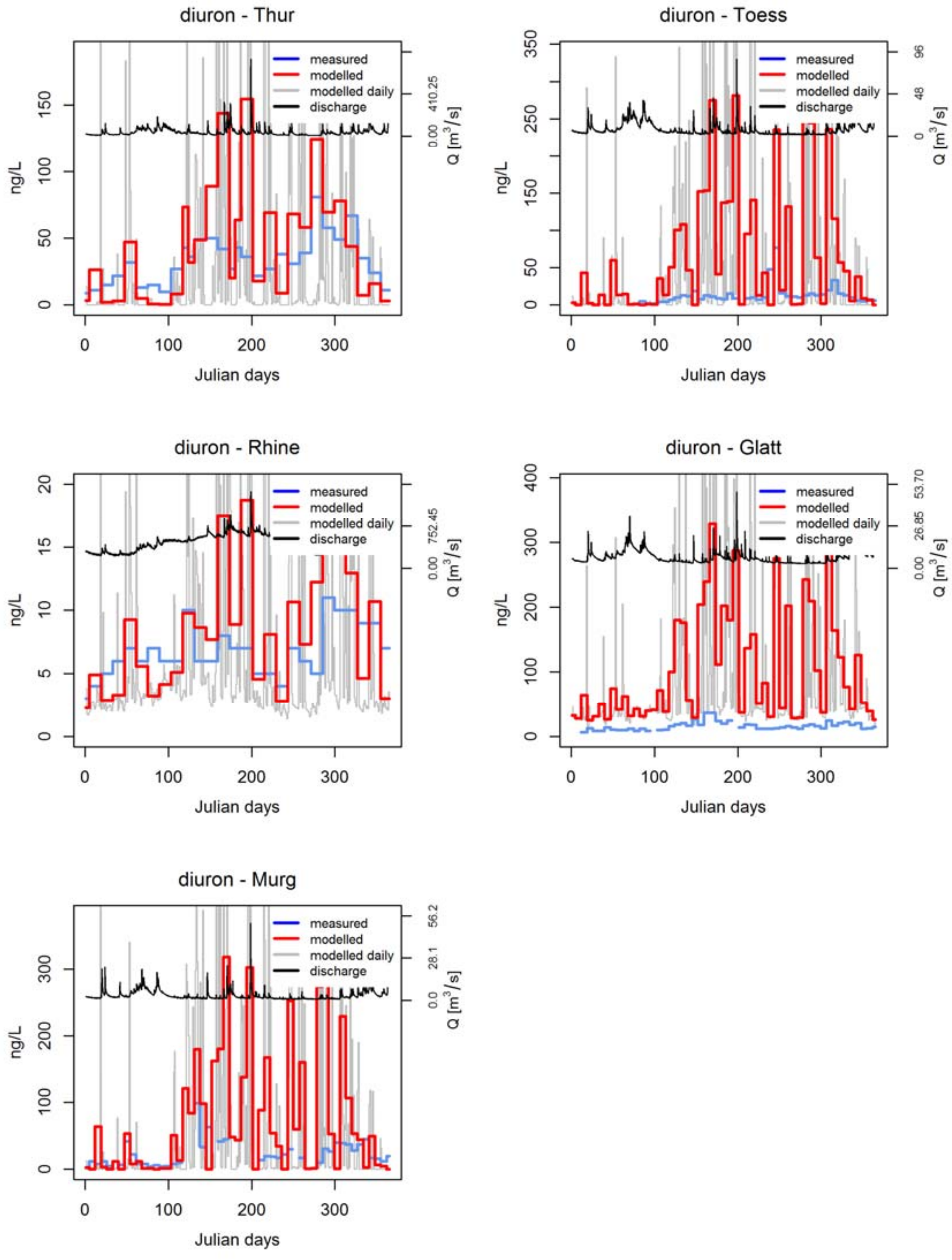


Figure S20 : Validation of diuron at the NADUF catchments with the calibrated parameter set from Mönchaltdorf. Note that the axis differ for the sites.

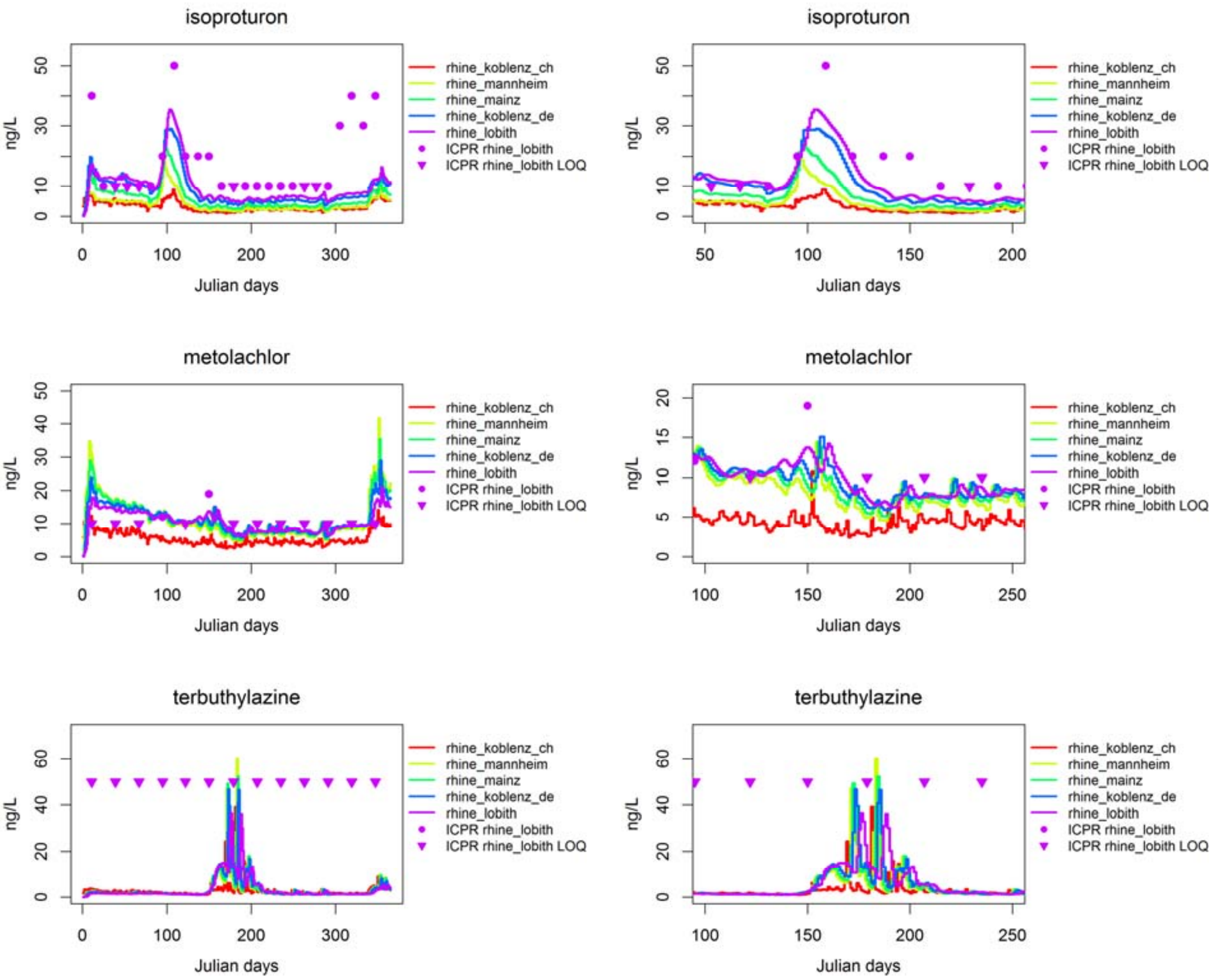


Figure S21 : Modelled concentrations at different points along the Rhine. Note the different axis for metolachlor in the right hand column.

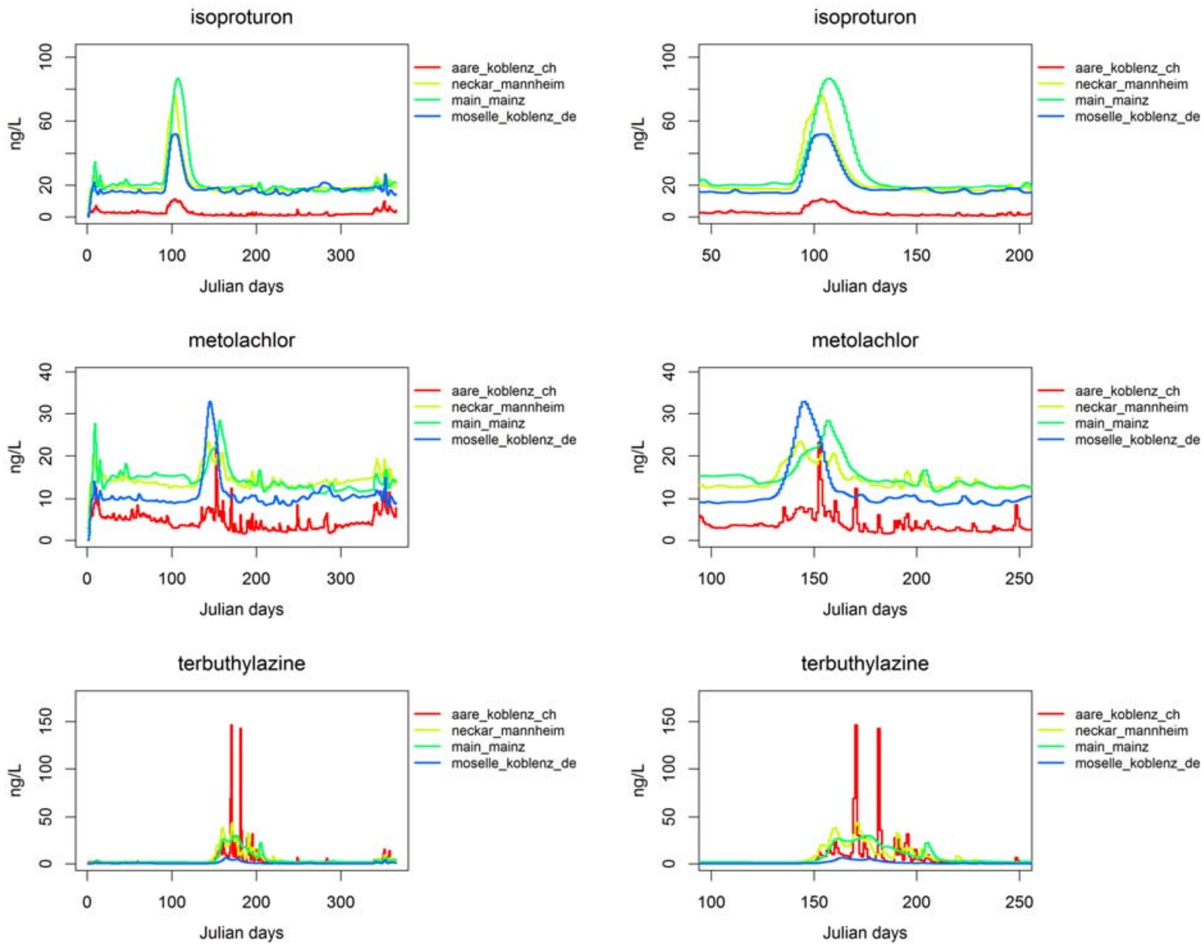


Figure S22 : Modelled isoproturon concentrations at the outlet of the main tributaries to the Rhine.

Appendix A12: Cumulative distributions of observed and calibrated concentrations

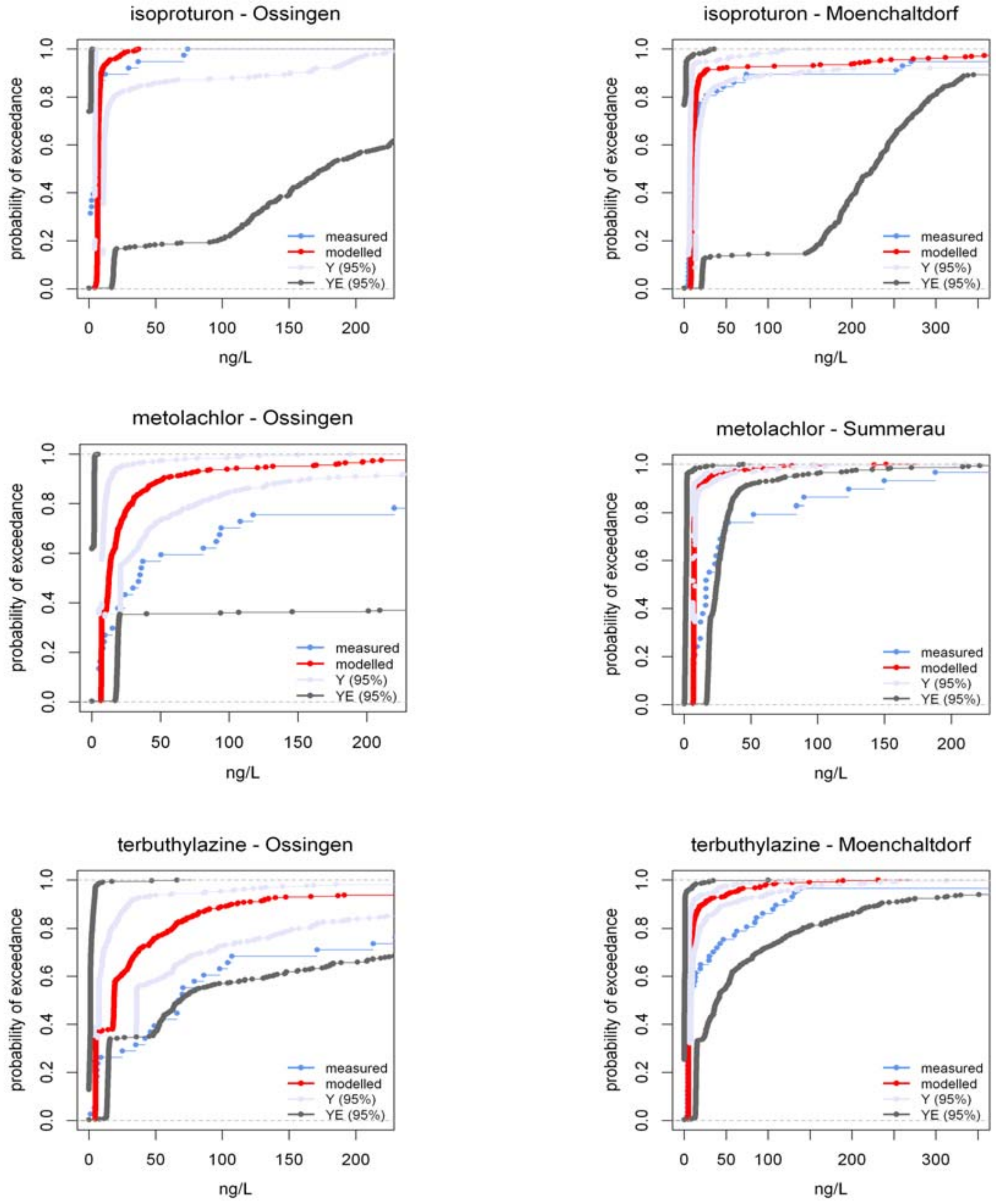


Figure S23 : Cumulative distributions of measured and modelled herbicide concentrations at the calibration sites. Y: parameter uncertainty prediction (95%), YE: parameter uncertainty and structural model error predictions (95%)

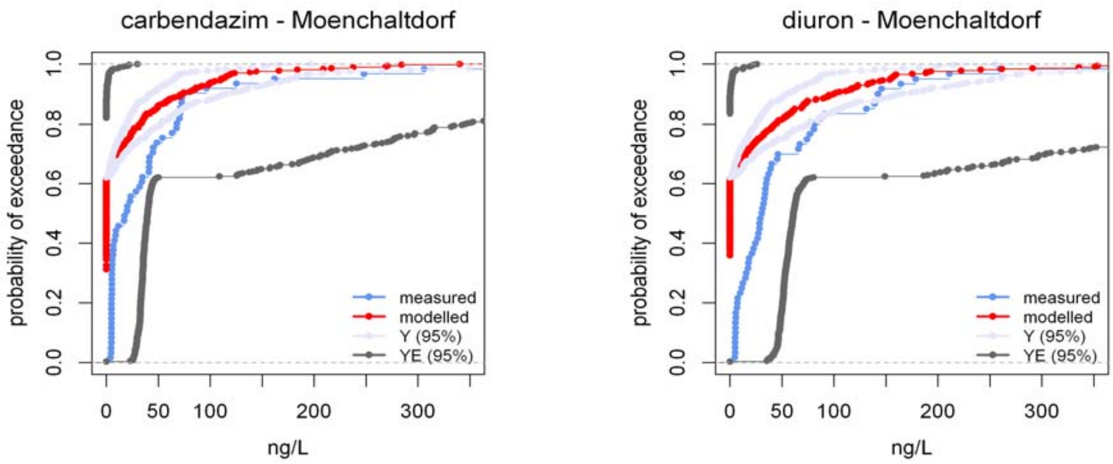


Figure S24 : Cumulative distributions of measured and modelled biocide concentrations at the calibration sites. Y: parameter uncertainty prediction (95%), YE: parameter uncertainty and structural model error predictions (95%).

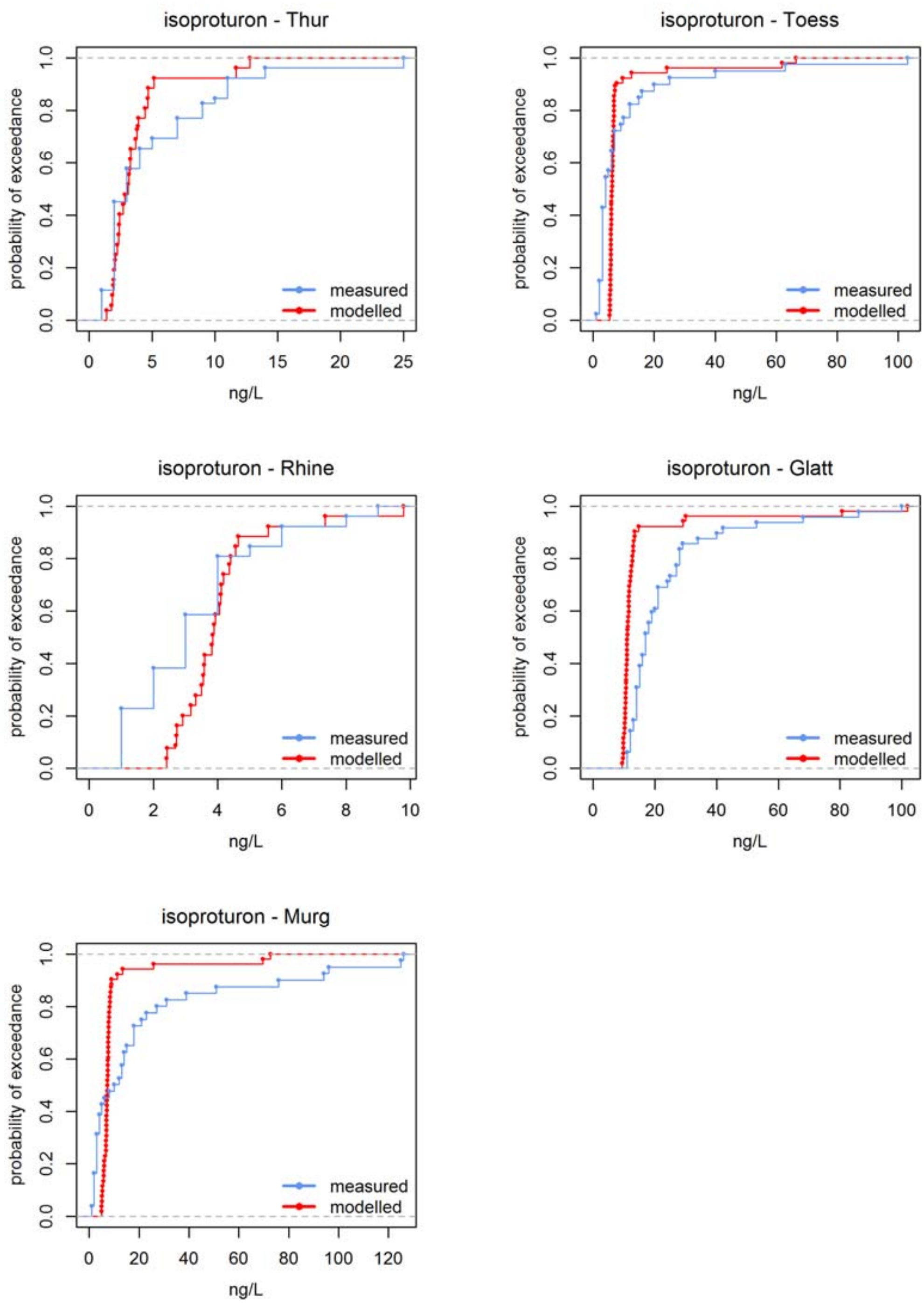


Figure S25 : **C**umulative distributions of measured and modelled isoproturon concentrations at the NADUF sites. Calibrated parameters from the Mönchaltdorf data set.

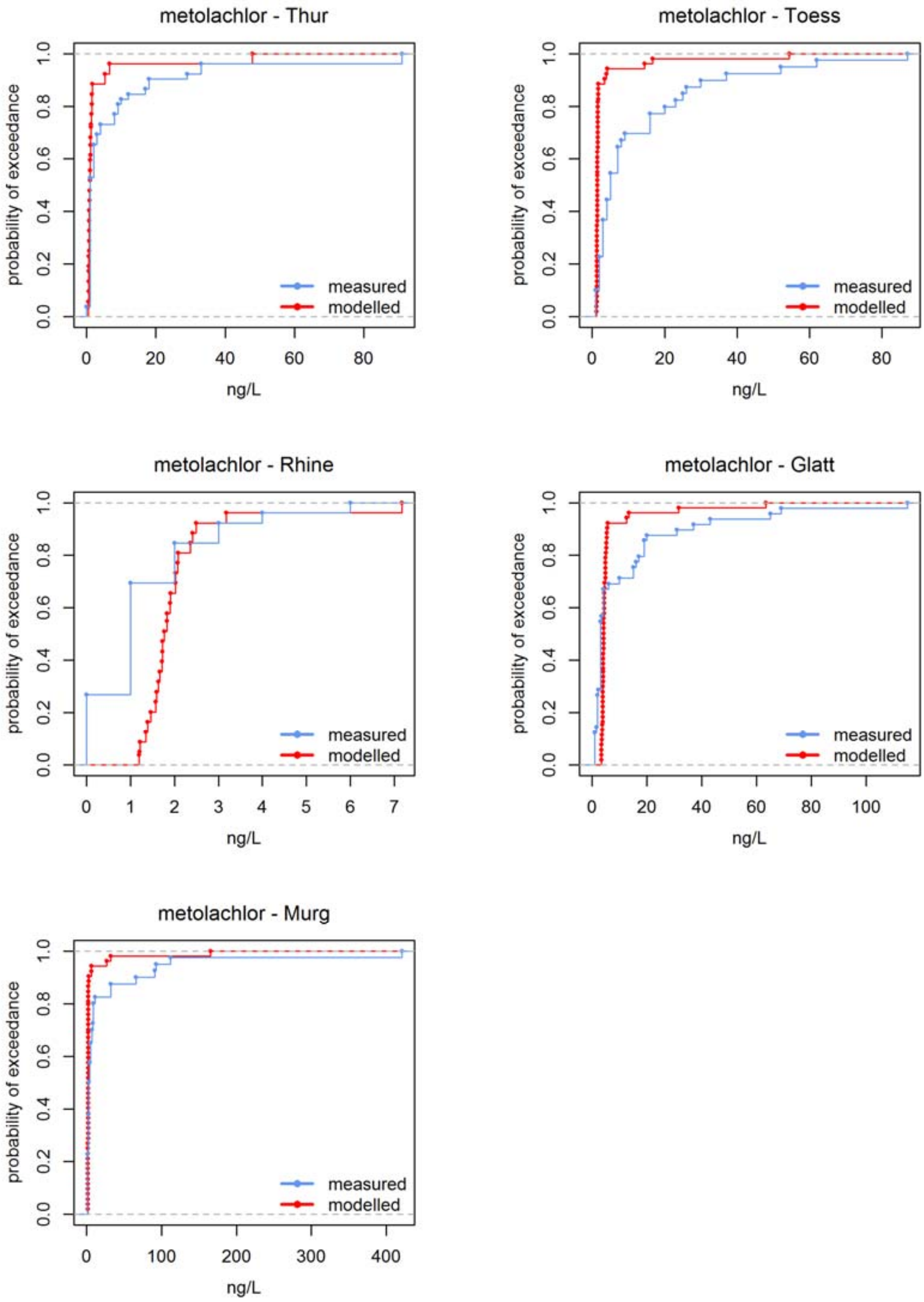


Figure S26 : **C**umulative distributions of measured and modelled metolachlor concentrations at the NADUF sites. Calibrated parameters from the Summerau data set.

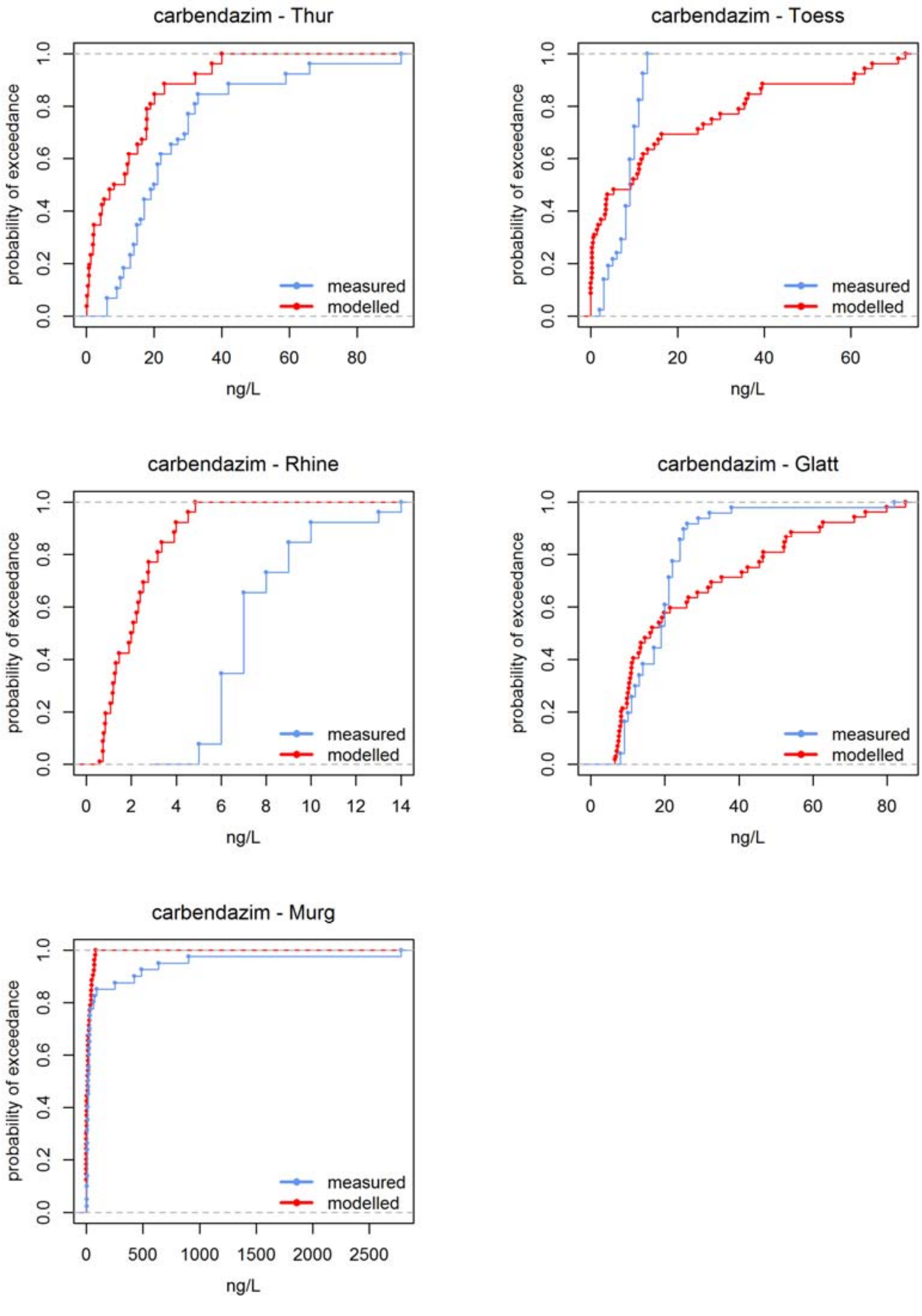


Figure S27 : **C**umulative distributions of measured and modelled carbendazim concentrations at the NADUF sites. Calibrated parameters from the Mönchaltdorf data set.

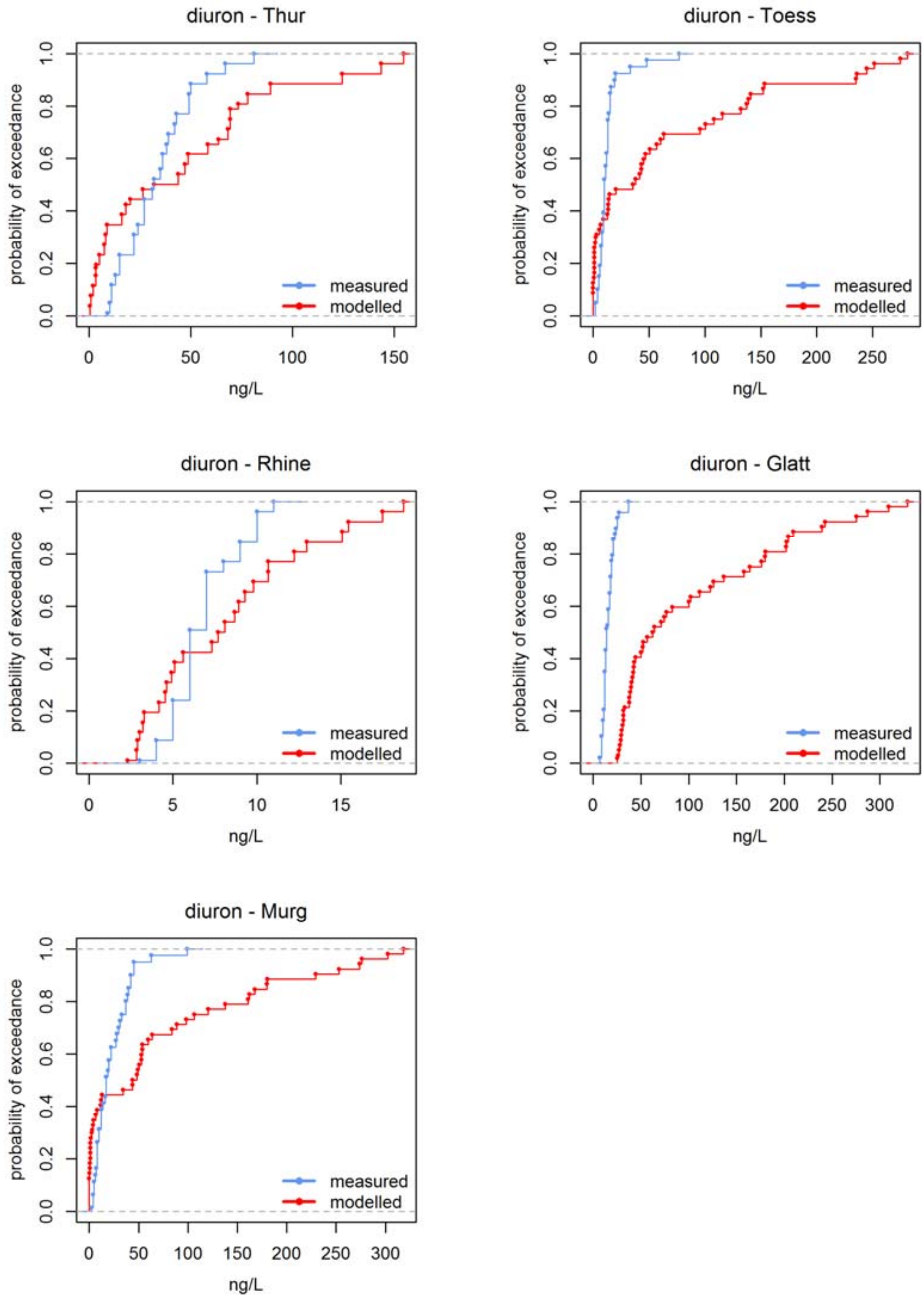


Figure S28 : **C**umulative distributions of measured and modelled diuron concentrations at the NADUF sites. Calibrated parameters from the Mönchaltdorf data set.

Appendix A13: Performance metrics

Table S8 Relative RMSE (RRMSE) during calibration for the different compound-site combinations.

	Mönchaltdorf	Ossingen	Summerau
Isoproturon	0.69	0.87	-
Metolachlor	-	0.62	0.63
Terbuthylazin	1.02	0.88	-
Carbendazim	0.95	-	-
Diuron	1.01	-	-

Table S9 : Percentage of modelled concentrations within the prediction intervals. moe – Mönchaltdorf, oss – Ossingen, sum – Summerau.

	Isoproturon		**S-metolachlor**		**Terbuthylazine**		**Carbendazim**	**Diuron**
	moe	oss	oss	summ	moe	oss	moe	moe
Furtbach	100	89	23	34	28	9	67	17
Salmsacher Aach	76	100	70	70	30	39	28	0
Surb	72	94	59	42	80	20	42	6
Limpach	100	100	68	23	67	100	19	0
Mentue	77	100	53	65	49	41	38	0

Table S10: Overview about the performance metrics for the different simulations.

Level	Sub-level	Routing	TestSite	Calibration Site	Compound	Cmax_obs [ng/L]	Cmax_mod [ng/L]	Cmax_rel	Fold difference	NSE	Cor	GRI	GRI_sorted	PBIAS	RRMSE
Calibration	Calibration	Load aggregation	Mönch	Mönch	Isoproturon	623	541	0.87	1.15	0.51	0.73	1.93	1.59	-31.1	0.69
Calibration	Calibration	Load aggregation	Mönch	Mönch	Terbuthylazin	697	334	0.48	2.09	-0.05	0.21	2.38	1.37	-19.7	1.02
Calibration	Calibration	Load aggregation	Mönch	Mönch	Carbendazim	306	116	0.38	2.63	0.08	0.37	5.01	3.46	-37.2	0.95
Calibration	Calibration	Load aggregation	Mönch	Mönch	Diuron	461	156	0.34	2.96	-0.05	0.3	5.47	3.85	-34.3	1.01
Calibration	Calibration	Load aggregation	Ossingen	Ossingen	Isoproturon	74	20	0.27	3.76	0.22	0.56	3.11	2.74	-9	0.87
Calibration	Calibration	Load aggregation	Ossingen	Ossingen	Metolachlor	2491	1180	0.47	2.11	0.61	0.85	2.05	1.37	-25.1	0.62
Calibration	Calibration	Load aggregation	Ossingen	Ossingen	Terbuthylazin	8072	1393	0.17	5.8	0.2	0.62	2.59	1.88	-49.9	0.88
Calibration	Calibration	Load aggregation	Summerau	Summerau	Metolachlor	346	151	0.44	2.29	0.59	0.85	1.82	1.41	-19.3	0.63
Validation	NAWA SPEZ	Load aggregation	Furtbach	Mönch	Isoproturon	350	372	1.06	1.06	-0.62	0.58	4.04	3.8	155.7	1.21
Validation	NAWA SPEZ	Load aggregation	Furtbach	Mönch	Terbuthylazin	340	1806	5.31	5.31	-33.14	-0.36	10.55	4.58	296.1	5.41
Validation	NAWA SPEZ	Load aggregation	Furtbach	Mönch	Carbendazim	55	89	1.61	1.61	-5.02	0.44	2.58	2.23	41.9	2.34
Validation	NAWA SPEZ	Load aggregation	Furtbach	Mönch	Diuron	52	424	8.15	8.15	-358.88	0.52	5.55	5.56	472.6	18.09
Validation	NAWA SPEZ	Load aggregation	Limpach	Mönch	Isoproturon	240	487	2.03	2.03	-0.74	0.98	1.93	1.9	75.9	1.26
Validation	NAWA SPEZ	Load aggregation	Limpach	Mönch	Terbuthylazin	490	573	1.17	1.17	0.09	0.77	2.52	2.47	42.1	0.85
Validation	NAWA SPEZ	Load aggregation	Limpach	Mönch	Carbendazim	33	14	0.43	2.33	0.06	0.72	2.42	2.42	-6.9	0.79
Validation	NAWA SPEZ	Load aggregation	Limpach	Mönch	Diuron	10	287	28.72	28.72	-2492.82	0.62	21.46	24.09	2584.8	46.71
Validation	NAWA SPEZ	Load aggregation	Mentue	Mönch	Isoproturon	41	1027	25.05	25.05	-834.03	0.9	7.51	7.64	1948	27.4
Validation	NAWA SPEZ	Load aggregation	Mentue	Mönch	Terbuthylazin	270	385	1.43	1.43	-1.78	0.7	2.52	2.48	93.3	1.52
Validation	NAWA SPEZ	Load aggregation	Mentue	Mönch	Carbendazim	16	30	1.84	1.84	-9.06	-0.38	3.64	2.7	16.1	2.99
Validation	NAWA SPEZ	Load aggregation	Mentue	Mönch	Diuron	30	141	4.7	4.7	-51.7	0.69	9.79	10.98	784.8	6.63
Validation	NAWA SPEZ	Load aggregation	Salmsacher Aach	Mönch	Isoproturon	56	120	2.14	2.14	-10.03	0.01	5.44	4.04	239	3.17
Validation	NAWA SPEZ	Load aggregation	Salmsacher Aach	Mönch	Terbuthylazin	130	2597	19.98	19.98	-489.72	0.72	4.46	3.77	691.2	20.88
Validation	NAWA SPEZ	Load aggregation	Salmsacher Aach	Mönch	Carbendazim	43	141	3.29	3.29	-20.17	0.27	3.78	3.62	260.8	4.34
Validation	NAWA SPEZ	Load aggregation	Salmsacher Aach	Mönch	Diuron	52	677	13.01	13.01	-387.43	0.49	12.65	17.07	1434.5	18.79
Validation	NAWA SPEZ	Load aggregation	Surb	Mönch	Isoproturon	220	490	2.23	2.23	-10.25	-0.02	3.69	2.35	219.6	3.2
Validation	NAWA SPEZ	Load aggregation	Surb	Mönch	Terbuthylazin	630	981	1.56	1.56	-0.56	0.9	2.62	2.62	105.1	1.14
Validation	NAWA SPEZ	Load aggregation	Surb	Mönch	Carbendazim	65	44	0.68	1.47	-1.87	-0.41	3.61	2.67	39.5	1.61
Validation	NAWA SPEZ	Load aggregation	Surb	Mönch	Diuron	22	211	9.59	9.59	-513.61	0.54	9.84	10.26	1079.4	21.63
Validation	NAWA SPEZ	Load aggregation	Furtbach	Ossingen	Isoproturon	350	30	0.09	11.73	-0.12	-0.02	2.84	1.92	-71.9	1
Validation	NAWA SPEZ	Load aggregation	Furtbach	Ossingen	Metolachlor	790	3111	3.94	3.94	-20.18	-0.26	13.19	5.41	165.4	4.39
Validation	NAWA SPEZ	Load aggregation	Furtbach	Ossingen	Terbuthylazin	340	3426	10.08	10.08	-114.69	-0.35	11.63	5.47	612.4	9.96
Validation	NAWA SPEZ	Load aggregation	Limpach	Ossingen	Isoproturon	240	31	0.13	7.83	-0.16	0.76	3.14	3.16	-78.1	1.03
Validation	NAWA SPEZ	Load aggregation	Limpach	Ossingen	Metolachlor	530	1199	2.26	2.26	-4.47	0.84	4.14	3.55	93.6	2.23
Validation	NAWA SPEZ	Load aggregation	Limpach	Ossingen	Terbuthylazin	490	1564	3.19	3.19	-16.55	0.76	4.41	4.42	272.5	3.75
Validation	NAWA SPEZ	Load aggregation	Mentue	Ossingen	Isoproturon	41	56	1.38	1.38	0.78	0.98	1.97	1.96	41.8	0.44
Validation	NAWA SPEZ	Load aggregation	Mentue	Ossingen	Metolachlor	160	758	4.74	4.74	-61.76	0.56	4.44	4.13	205.4	7.52
Validation	NAWA SPEZ	Load aggregation	Mentue	Ossingen	Terbuthylazin	270	876	3.24	3.24	-29.68	0.45	4.41	4.49	358.2	5.05
Validation	NAWA SPEZ	Load aggregation	Salmsacher Aach	Ossingen	Isoproturon	56	16	0.29	3.45	-0.06	0.02	3.21	2.71	-23.4	0.98
Validation	NAWA SPEZ	Load aggregation	Salmsacher Aach	Ossingen	Metolachlor	250	3082	12.33	12.33	-141.69	0.63	4.72	3.89	452.9	11.39
Validation	NAWA SPEZ	Load aggregation	Salmsacher Aach	Ossingen	Terbuthylazin	130	2215	17.04	17.04	-324.3	0.68	4.82	4.25	562.9	17
Validation	NAWA SPEZ	Load aggregation	Surb	Ossingen	Isoproturon	220	27	0.12	8.18	-0.14	-0.01	2.05	2.03	-68.8	1.02
Validation	NAWA SPEZ	Load aggregation	Surb	Ossingen	Metolachlor	1100	1752	1.59	1.59	-2.09	0.18	5.9	4.14	6.3	1.67
Validation	NAWA SPEZ	Load aggregation	Surb	Ossingen	Terbuthylazin	630	2974	4.72	4.72	-25.43	0.82	5.17	5.29	424.7	4.69
Validation	NAWA SPEZ	Load aggregation	Furtbach	Summerau	Metolachlor	790	711	0.9	1.11	-0.9	0.01	5.73	4.44	-36.5	1.31
Validation	NAWA SPEZ	Load aggregation	Limpach	Summerau	Metolachlor	530	454	0.86	1.17	-1.52	0.03	6.08	3.59	-14.5	1.51
Validation	NAWA SPEZ	Load aggregation	Mentue	Summerau	Metolachlor	160	232	1.45	1.45	-2.65	0.4	3.94	3.45	-4.1	1.81
Validation	NAWA SPEZ	Load aggregation	Salmsacher Aach	Summerau	Metolachlor	250	292	1.17	1.17	-1.34	-0.07	3.78	2.56	-14.5	1.46
Validation	NAWA SPEZ	Load aggregation	Surb	Summerau	Metolachlor	1100	517	0.47	2.13	-0.58	0.52	6.11	6.62	-70.4	1.19

Table S10 (cont.): Overview about the performance metrics for the different simulations.

Level	Sub-level	Routing	TestSite	Calibration Site	Compound	Cmax_obs [ng/L]	Cmax_mod [ng/L]	Cmax_rel	Fold difference	NSE	Cor	GRI	GRI_sorted	PBIAS	RRMSE
Validation	NADUF	Load aggregation	Thur	Mönch	Isoproturon	14	14	1.01	1.01	-0.08	0.3	2.29	1.49	-32.2	1.04
Validation	NADUF	Load aggregation	Thur	Summerau	Metolachlor	91	70	0.77	1.3	0.75	0.92	3.17	2.82	-54.5	0.5
Validation	NADUF	Load aggregation	Thur	Mönch	Carbendazim	93	32	0.34	2.95	-0.57	0.49	4.51	4.57	-67.9	1.25
Validation	NADUF	Load aggregation	Thur	Mönch	Diuron	81	122	1.51	1.51	-0.16	0.77	2.78	2.52	-4.5	1.08
Validation	NADUF	Load aggregation	Rhine-Rekingen	Mönch	Isoproturon	8	10	1.25	1.25	0.26	0.64	1.81	1.68	23.7	0.86
Validation	NADUF	Load aggregation	Rhine-Rekingen	Summerau	Metolachlor	6	8	1.27	1.27	0.29	0.76	3.36	3.48	54.2	0.84
Validation	NADUF	Load aggregation	Rhine-Rekingen	Mönch	Carbendazim	14	5	0.34	2.95	-6.09	0.56	4.05	4.03	-72	2.66
Validation	NADUF	Load aggregation	Rhine-Rekingen	Mönch	Diuron	11	18	1.67	1.67	-3.4	0.58	1.6	1.39	20.6	2.1
Validation	NADUF	Load aggregation	Murg	Mönch	Isoproturon	126	71	0.57	1.76	0.14	0.53	3	2.53	-52.3	0.92
Validation	NADUF	Load aggregation	Murg	Summerau	Metolachlor	421	304	0.72	1.39	0.77	0.93	2.94	2.64	-56.6	0.48
Validation	NADUF	Load aggregation	Murg	Mönch	Carbendazim	2779	70	0.03	39.73	-0.1	-0.06	6.19	6.09	-89.6	1.05
Validation	NADUF	Load aggregation	Murg	Mönch	Diuron	99	271	2.74	2.74	-18.37	0.35	5.12	4.37	164.1	4.4
Validation	NADUF	Load aggregation	Töss	Mönch	Isoproturon	103	65	0.63	1.58	0.44	0.66	2.26	1.87	-10.3	0.74
Validation	NADUF	Load aggregation	Töss	Summerau	Metolachlor	87	100	1.15	1.15	0.35	0.75	3.54	3.57	-63.1	0.81
Validation	NADUF	Load aggregation	Töss	Mönch	Carbendazim	13	65	5	5	-42	0.52	4.06	3.83	125.4	6.56
Validation	NADUF	Load aggregation	Töss	Mönch	Diuron	77	252	3.27	3.27	-49.76	0.36	6.27	5.53	449.4	7.12
Validation	NADUF	Load aggregation	Glatt	Mönch	Isoproturon	53	102	1.92	1.92	-0.42	0.21	1.93	1.77	-43.9	1.19
Validation	NADUF	Load aggregation	Glatt	Summerau	Metolachlor	115	88	0.77	1.3	0.03	0.38	2.51	2.29	-44.1	0.98
Validation	NADUF	Load aggregation	Glatt	Mönch	Carbendazim	82	71	0.87	1.15	-2.06	0.39	1.91	1.49	34.4	1.75
Validation	NADUF	Load aggregation	Glatt	Mönch	Diuron	37	276	7.45	7.45	-296.14	0.63	4.77	4.77	513.6	17.24
Validation	IRMS	Aquasim	Basel	Mönch	Isoproturon	29	9	0.32	3.16	0.22	0.67	1.84	1.51	-37.2	0.88
Validation	IRMS	Aquasim	Basel	Summerau	Metolachlor	52	14	0.27	3.71	-0.82	0.28	2.36	2.16	-56.8	1.35
Validation	IRMS	Aquasim	Basel	Mönch	Terbuthylazin	37	70	1.9	1.9	-3.28	0.04	2.93	2.41	-29.5	2.1
Validation	IRMS	Aquasim	Basel	Mönch	Carbendazim	11	12	1.13	1.13	-8.04	0.27	2.74	2.59	-39.8	3.01
Validation	IRMS	Aquasim	Basel	Mönch	Diuron	14	44	3.14	3.14	-49.22	0.12	2.39	2.07	105.2	7.09
Validation	IRMS	Load aggregation	Basel	Mönch	Isoproturon	29	10	0.33	3.05	0.35	0.84	1.64	1.49	-35.2	0.81
Validation	IRMS	Load aggregation	Basel	Summerau	Metolachlor	52	21	0.4	2.51	-0.71	0.33	2.23	2.05	-54.2	1.31
Validation	IRMS	Load aggregation	Basel	Mönch	Terbuthylazin	37	173	4.68	4.68	-12.23	-0.02	2.16	1.54	6.7	3.64
Validation	IRMS	Load aggregation	Basel	Mönch	Carbendazim	11	37	3.39	3.39	-10.08	0.23	2.07	1.87	-21.6	3.33
Validation	IRMS	Load aggregation	Basel	Mönch	Diuron	14	144	10.31	10.31	-121.41	-0.04	2.52	2.34	161.3	11.06

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