Water Emerging Contaminants& Nanoplastics

Occurrence of pharmaceutically active compounds, parabens and their main metabolites in soils amended with sludge and compost

SOIL AND SLUDGE CHARACTERIZATION

The soil was characterised by the determination of the texture, pH, electrical conductivity (EC), organic matter content (OM), cationic exchange capacity (CEC), and exchangeable cations content (Ca^{2+}, Mg^{2+}, K^{+}, and Na^{+}). The texture was analysed using Bouyoucos method (Beretta et al., 2014). Electrical conductivity (EC) and pH were measured in soil:water solution (1:2.5 w/v and 1:5, w/v, respectively). OM was determined by dichromate oxidation (Nelson and Sommers, 1982). CEC was determined by shaking the soil with 1 N sodium acetate, then leaching with 96% ethanol, and finally shaking and leaching three times with 1N ammonium acetate. Sodium concentration in the extract was measured with a flame photometer (Meimaroglou and Mouzakis, 2019). For the determination of exchangeable cations samples were treated with 1N ammonium acetate solution and the extracts were analysed using a inductively coupled plasma (ICP)-optical emission spectrometer (Yuan et al., 2007). Supplementary Table 1 shows the characteristics of the studied soil. Sludge and compost were characterized by the determination of their pH and OM content. Both parameters were determined following the same procedures described above for the soil.

Supplementary Table 1. Characteristics of soil, sludge and compost used for dissipation experiments

Property, units	Soil	Sludge	Compost
Fine sand, wt.%	14.0	n.a.	n.a.
Coarse sand, wt.%	51.6	n.a.	n.a.
Silt, wt.%	18.4	n.a.	n.a.
Clay, wt.%	16.0	n.a.	n.a.
pH	8.06	5.85	6.33
EC, µS cm^{-1}	126	n.a.	n.a.
OM, wt.%	2.01	48.5	27.3
CEC, meq kg^{-1}	144	n.a.	n.a.
Exchangeable Ca, meq kg^{-1}	106	n.a.	n.a.
Exchangeable Mg, meq kg^{-1}	19.5	n.a.	n.a.
Exchangeable K, meq kg^{-1}	4.6	n.a.	n.a.
Exchangeable Na, meq kg^{-1}	<0.5	n.a.	n.a.

Wt: weight

ANALYSIS OF TARGET COMPOUNDS

Target compounds were analyzed using a previously reported and validated analytical method (Malvar et al., 2020). The method was based on sample treatment by selective pressurized liquid extraction (SPLE) and determination by liquid chromatography-tandem mass spectrometry. For sample treatment, 2 g of lyophilized solid (sludge, compost, sludge-amended soil or compost-amended soil) were mixed with 1 g of diatomaceous earth. The 10 mL stainless steel cells were fixed as follow: first a cellulose filter (Thermo 068093) was placed at the bottom of the cell; then diatomaceous earth (0.3 g), clean-up sorbents (0.4 g of PSA and 0.8 g of C18), sample mixed with diatomaceous earth and, diatomaceous earth to fill the cell volume were added to the cells. Another cellulose filter was placed at the top of the cell. The conditions applied for SPLE were: methanol (MeOH) containing 0.5% v/v of formic acid as extraction solvent, 5 min of pre-heating time, extraction pressure of 1500 psi, extraction temperature of 50 °C, 2 extraction cycles with a static time of 5 min each, a flush volume of 60% of the cell volume and a nitrogen purge of 2 min. The extracts were transferred to clean tubes, evaporated to dryness using a gentle nitrogen stream (XcelVap® system), reconstituted in 0.5 mL of MeOH:water mixture (50:50, v/v), filtered through a 0.22 µm cellulose syringe filter, and collected into automatic injector vial for injection (10 µL) into the LC-MS/MS system.

The instrument was an Agilent 1200 series HPLC coupled to a 6410 triple quadrupole (QqQ) mass spectrometer (MS) with an electrospray ionization (ESI) source. Chromatographic separation was carried out using a Kinetex® polar C18 column (50 mm x 3 mm i.d., 2.6 µm particle size) (Phenomenex, Torrance CA) protected by a SecurityGuard™ ULTRA C18 guard column (2mm x 3mm i.d.) (Phenomenex, Torrance CA). The mobile phase was composed of water (0.1% v/v, formic acid) (solvent A) and MeOH (0.1% v/v, formic acid) (solvent B). The column was thermostated at 35 ºC. Flow-rate was 0.6 mL/min. Gradient elution was as follows: linear increase of solvent B from 2% to 38% in 9.5 min (held for 2.5 min), then to 50% in 3.5 min (held for 2.5 min) and, finally, to 98% in 4 min (held for 1 min). Back to initial conditions was carried out by linear decrease of solvent B from 98% to 2% in 2 min, held for 5 min for re-equilibration.

MS/MS determination was performed by applying the following parameters: capillary voltage, 4000 V; drying-gas flow rate, 9 L/min; drying-gas temperature, 350°C; nebulizer pressure, 40 psi. The determination was done using multiple reaction monitoring (MRM) mode. For each compound, two transitions were selected. The most abundant one (MRM1) was used the quantification. The less abundant transition (MRM2), and the relations between both transitions, were used for identification. The transitions, fragmentors and collision energies are showed in Table 2.

The analytical method was validated by the determination of matrix effect, accuracy, precision, linearity, method detection limits (MDL) and method quantitation limits (MQL). More details about the determination of these analytical parameters can be found in Malvar et al., 2020. Briefly, matrix effects were in the range from 9 to 135%. Because of the high suppression affecting some of the studied compounds, matrix-matched calibration were used for quantification. Accuracy, calcualted at four spike concentration levels, was in the ranges 60-120, 60-121, 59-125 % in soil, compost and digested sludge, respectively. Precision was lower than 21 % for all types of samples. Method detection and quantification limits were lower than 4.31 µg kg^{-1} and 14.4 µg kg^{-1} in all types of samples. Matrix-matched calibration curves were linear in the range from instrumental method quantification limits to 1000 µg L^{-1} for all the compounds.

Supplementary Table 2. LC-MS/MS parameters.

Compound	Ionisation mode	Precursor Ion (*m/z*)	Product ions (MRM1/MRM2) (*m/z*)	Fragmentor (V)	CE (V)	Retention time (min)	Ion ratio
SMX	Positive	254.1	92.1/65.0	80	28/40	6.8	58.2
AcSMX	Positive	296.1	65.0/134.0	60	40/20	9.2	68.6
DIC	Positive	296.0	214.0/250.0	60	32/8	21.1	37.8
4-OH-DIC	Positive	312.0	230.0/266.0	60	40/8	17.3	46.1
CBZ	Positive	237.1	194.1/179.0	140	16/36	13.2	13.6
3-OH-CBZ	Positive	253.1	210.1/167.0	120	16/40	11.4	18.5
10-OH-CBZ	Positive	255.1	194.1/237.1	60	20/4	10.3	93.4
EP-CBZ	Positive	253.1	180.1/236.1	60	24/4	10.7	68.6
IBU	Positive	207.1	161.1/119.1	100	4/20	21.4	31.5
1-OH-IBU	Negative	221.1	159.1/177.1	60	4/0	14.2	65.3
2-OH-IBU	Negative	221.1	177.1	40	0	12.7	-
CBX-IBU	Negative	235.1	191.1/73.0	40	0/8	13.2	51.0
MeP	Negative	151.0	92.0/136.0	60	20/12	8.3	60.7
OH-MeP	Negative	167.0	108.0/152.0	100	20/12	6.4	22.3
PrP	Negative	179.1	92.0/136.0	60	24/12	14.2	30.8
3,4-DHB	Negative	153.0	109.0	80	12	2.3	-
4-HB	Negative	137.0	93.0/65.1	60	16/32	3.7	8.4

CE: collision energy; RT: retention time.

Supplementary Table 3. Concentrations and relative standard deviations of the target compounds in digested sludge and compost (n=5)

	Digested sludge			Compost		
	Range (μg/kg)	Mean (μg/kg)	RSD (%)	Range (μg/kg)	Mean (μg/kg)	RSD (%)
SMX	<MDL	<MDL	-	<MDL	<MDL	-
AcSMX	<MDL	<MDL	-	<MDL	<MDL	-
DIC	0.71-1.09	0.90	12	<MDL	<MDL	-
4-OH-DIC	1.78-1.97	1.86	3	<MDL	<MDL	-
CBZ	0.52-0.83	0.63	16	0.44-1.95	0.67	56
3-OH-CBZ	<MDL	<MDL	-	<MDL	<MDL	-
10-OH-CBZ	2.36-2.80	5.54	5	<MDL	<MDL	-
EP-CBZ	<MDL	<MDL	-	<MDL	<MDL	-
IBU	<MDL	<MDL	-	<MDL	<MDL	-
1-OH-IBU	<MDL	<MDL	-	<MDL	<MDL	-
2-OH-IBU	<MDL	<MDL	-	<MDL	<MDL	-
CBX-IBU	4.37-5.52	4.95	8	<MDL	<MDL	-
MeP	16.7-29.0	22.1	17	5.30-13.9	9.37	28
OH-MeP	9.18-12.1	10.9	8	9.05-11.1	9.73	6
PrP	<MDL	<MDL	-	<MDL	<MDL	-
3,4-DHB	13.0-15.8	14.1	6	28.6-42.0	36.4	12
4-HB	95.4-126	112	9	45.6-60.8	52.2	8

Supplementary Table 4. Optimized parameters of the kinetic models applied to the degradation of PPCs and their metabolites in sludge-amended soil. MSE: mean square error of the applied model. $R^{2:}$ determination coefficient of the measured concentration *vs* the concentration obtained with the model.

		SMZ	AcSMZ	DIC	4-OH-DIC	CBZ	3-OH-CBZ	10-OH-CBZ	EP-CBZ
SFO	k (day^{-1})	0.174	0.199	0.081	0.646	0.020	0.063	0.052	0.033
	C_0 (μg kg^{-1})	1060	587	229	438	633	658	663	570
	MSE	2226	636	816	191	14545	6107	5163	10911
	R^2	0.962	0.964	0.802	0.922	0.538	0.854	0.880	0.718
	Slope	0.97	0.95	0.90	0.94	0.50	0.81	0.82	0.67
	DT50 (day)	3.98	3.48	8.56	1.07	33.0	11.0	13.3	21.0
	DT90 (day)	13.2	11.6	28.4	3.56	110	36.5	44.3	69.8
BEM	k_1 (day^{-1})	0.170	0.199	0.081	0.395	0.020	0.063	0.052	0.013
	k_2 (kg $μg^{-1}$ day^{-1})	$1.53x10^{-6}$	0	$9.39x10^{-5}$	0.001	0	0	0	$4.88x10^{-5}$
	C_0 (μg kg^{-1})	1060	587	255	438	633	658	663	570
	MSE	2244	636	814	176	14545	6107	5163	12571
	R^2	0.962	0.964	0.822	0.926	0.538	0.854	0.880	0.676
	Slope	0.97	0.95	0.97	0.91	0.50	0.81	0.82	0.57
	DT50 (day)	4.05	3.48	7.06	0.98	33.0	11.0	13.3	21.3
	DT90 (day)	13.5	11.6	25.6	4.21	110	36.5	44.3	104
FODED	k_1 (day^{-1})	0.174	0.199	0	0.801	0.020	0.063	0.052	0.033
	k_2 (day^{-1})	0.174	0.199	0.132	0.18	0.020	0.063	0.052	0.033
	C_{sorb} (μg kg^{-1})	678	417	227	50.6	385	471	438	325
	C_{sol} (μg kg^{-1})	382	169	30.1	417	248	187	225	245
	MSE	2226	636	577	152	14545	6107	5163	10911
	R^2	0.962	0.964	0.843	0.935	0.538	0.854	0.880	0.718
	Slope	0.97	0.95	0.84	0.94	0.50	0.81	0.82	0.67
	DT50 (day)	3.98	3.48	6.32	0.97	33.0	11.0	13.3	21.0
	DT90 (day)	13.2	11.6	>1000	3.64	110	36.5	44.3	69.8
FOTC	k_1 (day^{-1})	0.174	0.199	0.117	0.646	0.020	0.063	0.049	0.033
	k_2 (day^{-1})	0.174	0.199	0	1.90	0.020	0.063	0.049	0.033
	k_r (day^{-1})	0.219	0.338	0.015	0	0.051	0.091	0.099	0.073
	C_0 (μg kg^{-1})	1060	587	257	438	633	658	579	570
	MSE	2226	636	577	191	14545	6107	6279	10911
	R^2	0.962	0.964	0.843	0.922	0.530	0.854	0.886	0.718
	Slope	0.97	0.95	0.84	0.94	0.53	0.81	0.71	0.67
	DT50 (day)	3.98	3.48	6.29	1.07	30.1	11.0	14.2	21.0
	DT90 (day)	13.2	11.6	312	3.56	100	36.5	47.0	69.8
MHSM	k (day^{-1})	0.174	0.199	0.081	0.646	0.020	0.063	0.052	0.033
	t_b (day)	0.16	1.06	0	0	8.74	1.121	2.23	4.20
	C_0 (μg kg^{-1})	1031	475	229	438	533	613	590	496
	MSE	2226	636	816	191	14545	6107	5163	10911
	R^2	0.962	0.964	0.802	0.922	0.538	0.854	0.880	0.718
	Slope	0.97	0.95	0.90	0.94	0.51	0.81	0.82	0.67

	DT50 (day)	4.14	4.54	8.56	1.07	39.6	12.1	15.6	25.2
	DT90 (day)	17.4	16.1	36.9	4.64	149	48.6	59.8	95.0
LM	a_{max}	0.169	0.290	0.081	0.646	0.067	0.107	0.457	0.073
	r	0.19	1.09	0.004	1.55	0.913	1.10	0.075	0.743
	a_0	0.169	0.008	0.081	0.646	10^{-12}	3.30×10^{-6}	0.016	5.26×10^{-7}
	C_0 (μg kg^{-1})	953	431	229	438	489	489	564	493
	MSE	26079	247	816	191	7344	3369	2789	7724
	R^2	0.962	0.985	0.802	0.922	0.683	0.919	0.933	0.853
	Slope	0.88	0.98	0.90	0.94	0.86	0.89	0.92	0.95
	DT50 (day)	4.10	5.62	8.56	1.07	37.6	15.9	19.9	25.4
	DT90 (day)	13.6	11.2	28.4	3.56	61.7	31.0	35.3	47.5

Supplementary Table 4. (Continuation)

		IBU	1-OH-IBU	2-OH-IBU	CBX-IBU	MeP	OH-MeP	PrP	3,4-DHB	4-HB
SFO	k	0.048	0.069	0.033	0.100	4.28	0.129	1.85	0.179	0.585
	C_0 (μg kg^{-1})	309	761	535	326	14812	322	1936	106	1017
	MSE	1365	16074	14127	14793	7.42	1342	7.17	268	1127
	R^2	0.823	0.770	0.512	0.290	0.997	0.823	0.998	0.673	0.928
	Slope	0.84	0.70	0.54	0.32	1.0	0.69	1.0	0.68	0.51
	DT50 (day)	14.4	10.0	21.0	6.93	0.16	5.37	0.37	3.87	1.18
	DT90 (day)	48.0	33.4	69.8	23.0	0.54	17.8	1.24	12.9	3.94
BEM	k_1 (day^{-1})	0.045	0.069	0.032	0.101	1.99	0.129	1.85	0.038	0.585
	k_2 (kg μg^{-1} day^{-1})	1.12x10^{-5}	0	1.53x10^{-6}	0	0.001	7.98x10^{-7}	5.23x10^{-7}	0.005	0
	C_0 (μg kg^{-1})	309	761	535	326	14812	322	1936	282	1017
	MSE	1364	16075	14128	14793	27	1342	7.17	255	1127
	R^2	0.823	0.770	0.512	0.290	0.982	0.823	0.998	0.689	0.928
	Slope	0.84	0.70	0.53	0.32	1.0	0.69	1.0	0.69	0.51
	DT50 (day)	14.6	10.0	21.3	6.86	0.06	5.37	0.37	0.68	1.18
	DT90 (day)	49.7	33.4	71.2	22.8	0.36	17.8	1.24	5.58	3.94
FODED	k_1 (day^{-1})	0.055	0.069	0	0	4.29	0.129	1.66	0.178	0.585
	k_2 (day^{-1})	0	0.069	0.048	0.119	0.016	0.129	3.21	0.182	0.585
	C_{sorb} (μg kg^{-1})	16.3	337	469	310	2.23	26.6	1239	77.8	112
	C_{sol} (μg kg^{-1})	299	424	86.4	22.4	14792	295	1736	27.9	905
	MSE	1350	16074	13826	14604	5.19	1342	7.56	268	1127
	R^2	0.824	0.770	0.0521	0.293	0.997	0.823	0.998	0.673	0.928
	Slope	0.82	0.70	0.52	0.29	1.0	0.69	1.0	0.68	0.51
	DT50 (day)	13.6	10.0	18.7	6.46	0.16	5.37	0.31	3.83	1.18
	DT90 (day)	54.1	33.4	>1000	28.2	0.54	17.8	1.13	12.7	3.94
FOTC	k_1 (day^{-1})	0.048	0.071	0.048	0.100	6.22	0.129	1.85	1.136	0.585
	k_2 (day^{-1})	0.048	0.071	0.048	0.100	3.90	0.129	1.85	0.134	0.585
	k_r (day^{-1})	4.88	0.130	0.264	0.184	5.05	0.234	1.41	0.370	2.068
	C_0 (μg kg^{-1})	309	670	535	326	14812	322	1930	274	1017
	MSE	1365	17898	17851	14793	7.54	1342	7.16	220	1127
	R^2	0.823	0.757	0.522	0.290	0.997	0.832	0.998	0.732	0.926
	Slope	0.84	0.61	0.60	0.32	1.0	0.82	1.0	0.72	0.93
	DT50 (day)	14.4	9.76	14.4	6.93	0.12	5.37	0.37	0.70	1.18
	DT90 (day)	48.0	32.4	48.0	23.0	0.50	17.8	1.24	7.40	3.94
MHSM	k (day^{-1})	0.048	0.069	0.033	0.100	4.54	0.129	1.85	0.179	0.585
	t_b (day)	0	0	0	0	0.06	0	0	0	2.08x10^{-6}
	C_0 (μg kg^{-1})	309	649	451	326	14812	322	1936	106	1017
	MSE	1365	18303	14127	14793	7.40	1342	7.17	268	1127
	R^2	0.823	0.760	0.567	0.290	0.997	0.832	0.998	0.673	0.926
	Slope	0.84	0.70	0.51	0.32	1.0	0.81	1.0	0.68	0.93
	DT50 (day)	14.4	12.4	26.1	6.93	0.21	5.37	0.37	3.87	1.18
	DT90 (day)	62.3	45.7	95.9	30.0	0.57	17.8	1.24	12.9	3.34
LM	a_{max}	0.050	0.137	0.126	104	4.28	0.129	1.85	0.169	1.83

r	2.64	1.26	1.03	1.23	3.93	10.2	4.40	0.029	5.93
a_0	4.42×10^{-6}	1.06×10^{-6}	7.00×10^{-7}	3.97×10^{-6}	4.28	4.83×10^{-6}	1.85	0.169	7.52×10^{-6}
C_0 (μg kg^{-1})	271	567	535	326	14810	284	1936	99.3	522
MSE	1340	8589	20209	12272	7.42	1342	7.17	270	116
R^2	0.826	0.871	0.467	0.469	0.996	0.832	0.998	0.673	0.993
Slope	0.84	0.83	0.63	0.69	1.0	0.80	1.0	0.65	1.0
DT50 (day)	17.4	14.4	17.2	9.98	0.16	6.37	0.37	4.10	2.45
DT90 (day)	49.6	26.2	30.0	11.0	0.54	18.8	1.24	13.6	3.35

Supplementary Table 5. Optimized parameters of the kinetic models applied to the degradation of PPCs and metabolites in compost-amended soil. MSE: mean square error of the applied model. R^2: determination coefficient of the measured concentration *vs* the concentration obtained with the model.

		SMZ	AcSMZ	DIC	4-OH-DIC	CBZ	3-OH-CBZ	10-OH-CBZ	EP-CBZ
SFO	k	0.137	0.305	0.167	0.653	0.024	0.156	0.110	0.027
	C_0 (μg kg^{-1})	686	699	340	279	613	638	614	489
	MSE	1158	127	292	48.3	8212	211	632	7632
	R^2	0.962	0.992	0.954	0.951	0.685	0.991	0.976	0.648
	Curve slope	1.0	1.0	0.97	0.97	0.66	1.0	0.96	0.59
	DT50 (day)	5.06	2.27	4.15	1.06	28.9	4.44	6.30	25.7
	DT90 (day)	16.8	7.55	13.8	3.53	95.9	14.8	20.9	85.3
BEM	k_1 (day^{-1})	0.124	0.305	0.167	0.485	0.024	0.136	0.109	0.027
	k_2 (kg μg^{-1} day^{-1})	3.23x10^{-5}	0	8.44x10^{-6}	0.001	0	5.56x10^{-5}	1.53x10^{-6}	0
	C_0 (μg kg^{-1})	686	699	342	279	613	638	614	489
	MSE	1147	127	292	45.5	8212	189	634	7632
	R^2	0.960	0.996	0.954	0.952	0.685	0.992	0.976	0.648
	Curve slope	0.99	1.0	0.97	0.95	0.66	0.99	0.96	0.59
	DT50 (day)	4.95	2.27	4.12	1.01	28.9	4.3	6.3	25.7
	DT90 (day)	17.4	7.55	13.8	3.93	95.9	15.4	21.1	85.3
FODED	k_1 (day^{-1})	0.159	0.305	0.178	0.286	0.024	0.086	0.110	0.027
	k_2 (day^{-1})	0.001	0.305	0	1.13	0.024	0.232	0.110	0.027
	C_{sorb} (μg kg^{-1})	26.5	418	5.38	299	352	483	510	324
	C_{sol} (μg kg^{-1})	687	282	341	68.7	261	195	104	166
	MSE	912	127	279	31.5	8212	135	632	7632
	R^2	0.966	0.996	0.955	0.966	0.685	0.994	0.976	0.648
	Curve slope	0.97	1.0	0.95	0.98	0.66	0.99	0.96	0.59
	DT50 (day)	4.60	2.27	3.98	0.75	28.9	3.83	6.30	25.7
	DT90 (day)	17.1	7.55	13.8	3.13	95.9	15.1	20.9	85.3
FOTC	k_1 (day^{-1})	0.121	0.305	0.172	0.653	0.024	0.156	0.110	0.029
	k_2 (day^{-1})	0.121	0.305	0	0.653	0.024	0.156	0.110	0.029
	k_r (day^{-1})	0.901	1.20	0.003	0	0.050	47.2	0.034	0.063
	C_0 (μg kg^{-1})	612	699	340	279	613	638	614	489
	MSE	1397	127	280	48.3	8212	211	632	7761
	R^2	0.952	0.992	0.954	0.951	0.685	0.991	0.976	0.641
	Curve slope	0.92	0.99	0.95	0.97	0.66	1.0	0.96	0.61
	DT50 (day)	5.73	2.27	4.06	2.99	28.9	4.33	6.30	23.9
	DT90 (day)	19.0	7.55	14.1	3.53	95.9	14.4	20.9	79.4
MHSM	k (day^{-1})	0.137	0.305	0.167	0.653	0.024	0.156	0.110	0.027
	t_b (day)	0.14	0.63	0	0	0	0	0.039	3.62
	C_0 (μg kg^{-1})	673	577	340	279	613	638	611	444
	MSE	1158	127	292	48.3	8212	211	632	7632
	R^2	0.962	0.992	0.954	0.951	0.685	0.991	0.976	0.648
	Curve slope	1.0	1.0	0.97	0.97	0.66	1.0	0.96	0.59

	DT50 (day)	5.20	2.90	4.15	1.06	28.9	4.44	6.34	29.3
	DT90 (day)	16.9	8.18	13.8	3.53	95.9	14.8	21.0	88.9
LM	a_{max}	0.169	0.341	0.160	0.653	0.067	0.127	0.110	0.072
	r	5.35	6.06	0.97	12.8	0.91	10.0	10.0	0.962
	a_0	0.0003	7.02×10^{-5}	0.160	0.653	5.26×10^{-7}	0.127	0.110	2.39×10^{-8}
	C_0 (μg kg^{-1})	661	499.2	333	279	571	536	614	489
	MSE	1446	79.9	295	49.8	12724	624	632	7445
	R^2	0.970	0.995	0.951	0.951	0.728	0.979	0.976	0.788
	Curve slope	1.0	1.0	0.99	0.97	1.0	0.89	0.96	1.1
	DT50 (day)	5.30	3.43	4.33	1.06	23.3	5.46	6.30	25.1
	DT90 (day)	14.8	8.15	14.4	3.53	47.3	18.1	20.9	47.5

Supplementary Table 5. (Continuation)

		IBU	1-OH-IBU	2-OH-IBU	CBX-IBU	MeP	OH-MeP	PrP	3,4-DHB	4-HB
SFO	k	0.143	0.229	0.042	0.204	1.87	0.309	1.24	2.35	0.448
	C_0 (μg kg^{-1})	393	814	594	471	1800	317	1202	1124	68.8
	MSE	179	1290	13953	4010	47.8	410	132	21.2	5.15
	R^2	0.980	0.957	0.641	0.722	0.990	0.885	0.985	0.964	0.898
	Curve slope	0.98	0.94	0.62	0.71	1.0	0.92	1.0	0.99	0.90
	DT50 (day)	14.4	10.0	21.0	6.93	0.37	2.24	0.56	0.29	1.55
	DT90 (day)	48.0	33.4	69.8	23.0	1.23	7.45	1.86	0.98	5.14
BEM	k_1 (day^{-1})	0.143	0.229	0.042	0.204	1.87	0.192	1.19	2.35	0.448
	k_2 (kg $μg^{-1}$ day^{-1})	0	0	0	0	0	0.001	6.80×10^{-5}	1.46×10^{-5}	0
	C_0 (μg kg^{-1})	393	814	594	471	1800	317	1202	1124	68.8
	MSE	179	1290	13954	4010	47.8	372	132	21.2	5.15
	R^2	0.980	0.957	0.641	0.722	0.990	0.890	0.984	0.964	0.898
	Curve slope	0.98	0.94	0.62	0.71	1.0	0.87	1.0	0.99	0.90
	DT50 (day)	4.85	3.03	16.5	3.42	0.37	1.67	0.56	0.29	1.55
	DT90 (day)	16.1	10.1	54.8	11.3	1.23	7.71	1.88	0.98	5.14
FODED	k_1 (day^{-1})	0.143	0.229	0.042	0.204	1.91	0.755	0.923	1.58	0.448
	k_2 (day^{-1})	0.143	0.229	0.042	0.204	0.040	0.162	2.25	2.96	0.448
	C_{sorb} (μg kg^{-1})	98.6	409	348	355.2	10.6	120	566	255	22.2
	C_{sol} (μg kg^{-1})	294	405	246	116.3	1798	331	1176	1044	46.5
	MSE	179	1290	13953	4010	21.5	280	116	20.8	5.15
	R^2	0.980	0.957	0.641	0.722	0.992	0.918	0.987	0.963	0.898
	Curve slope	0.98	0.94	0.62	0.71	0.99	0.94	1.0	0.99	0.90
	DT50 (day)	13.6	10.0	18.7	6.46	0.37	1.26	0.55	0.39	1.55
	DT90 (day)	54.1	33.4	>1000	28.2	1.23	6.41	2.10	1.34	5.14
FOTC	k_1 (day^{-1})	0.143	0.229	0.042	0.204	1.88	0.345	1.29	2.38	0.448
	k_2 (day^{-1})	0.143	0.229	0.042	0.204	1.85	0.184	1.07	2.34	0.448
	k_r (day^{-1})	0.097	0.189	0.053	0.076	1.73	0.168	0.358	3.09	0.079
	C_0 (μg kg^{-1})	393	814	594	471	1800	317	1202	1124	68.8
	MSE	179	1290	13953	4010	47.8	375	130	21.2	5.15
	R^2	0.980	0.957	0.641	0.722	0.990	0.957	0.984	0.964	0.898
	Curve slope	0.98	0.94	0.62	0.71	1.0	0.98	1.0	0.99	0.91
	DT50 (day)	4.85	3.03	16.5	3.40	0.37	2.19	0.55	0.29	1.55
	DT90 (day)	16.1	10.0	54.8	11.3	1.24	9.11	1.88	0.98	5.14
MHSM	k (day^{-1})	0.143	0.229	0.042	0.204	1.87	0.309	1.24	2.35	0.448
	t_b (day)	0.09	0.16	0	0.07	0	0	0	0	0
	C_0 (μg kg^{-1})	387	785	549	465	1800	317	1202	1124	68.7
	MSE	179	1290	13953	4010	47.8	410	132	21.2	5.15
	R^2	0.980	0.957	0.641	0.722	0.990	0.885	0.985	0.964	0.898
	Curve slope	0.98	0.94	0.62	0.71	1.0	0.93	1.0	0.99	0.90
	DT50 (day)	4.94	3.19	18.4	3.47	0.37	2.24	0.56	0.29	1.55
	DT90 (day)	16.2	10.2	56.7	11.4	1.23	7.45	1.86	0.98	5.14
LM	a_{max}	0.143	0.229	0.048	0.233	1.87	0.309	1.24	2.35	0.474

r	11.8	9.95	0.975	1.0	6.80	0.99	5.23	0.72	11.0
a_0	0.143	0.229	0.048	0.167	1.87	0.309	1.24	2.35	6.29×10^{-6}
C_0 (μg kg^{-1})	393	814	594	475	1800	317	1202	1123	44.9
MSE	179	1290	14669	4071	47.9	410	132	21.2	5.24
R^2	0.980	0.957	0.633	0.717	0.989	0.885	0.985	0.964	0.896
Curve slope	0.98	0.94	0.63	0.72	1.0	0.93	1.0	0.99	0.91
DT50 (day)	4.85	3.03	14.4	3.29	0.37	2.24	0.56	0.29	2.48
DT90 (day)	16.1	10.1	48.0	10.2	1.23	7.45	1.86	0.98	5.88

Supplementary Table 6. Optimized parameters of the kinetic models applied to the degradation of PPCs and metabolites in soil (experimental data from Malvar et al., 2020). MSE: mean square error of the applied model. R^2: determination coefficient of the measured concentration *vs* the concentration obtained with the model.

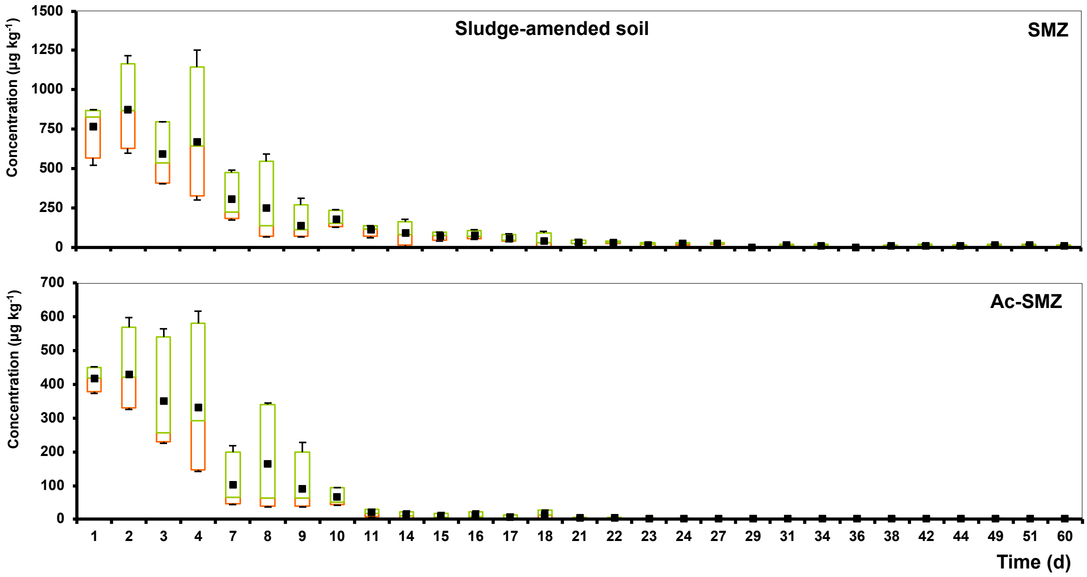
		SMZ	AcSMZ	DIC	4-OH-DIC	CBZ	3-OH-CBZ	10-OH-CBZ	EP-CBZ
SFO	k	0.113	0.578	0.104	0.611	0.003	0.167	0.013	0.009
	C_0 (µg kg^{-1})	367	408	611	291	459	575	124	429
	MSE	419	481	727	51.7	896	314	955	2378
	R^2	0.975	0.854	0.979	0.983	0.548	0.987	0.440	0.690
	Curve slope	1.0	0.74	1.0	1.0	0.56	1.0	0.42	0.72
	DT50 (day)	6.13	1.20	6.66	1.13	231	4.15	53.3	77.0
	DT90 (day)	20.4	3.98	22.1	3.77	768	13.8	177	256
BEM	k_1 (day^{-1})	0.120	0.578	0.084	0.164	0.003	0.149	0.013	0.009
	k_2 (kg µg^{-1} day^{-1})	0	0	4.99x10^{-5}	0.004	0	6.67x10^{-5}	1.53x10^{-6}	0
	C_0 (µg kg^{-1})	386	408	615	551	463	590	124	433
	MSE	436	480	676	24.6	907	309	954	2388
	R^2	0.975	0.854	0.997	0.983	0.550	0.986	0.420	0.692
	Curve slope	1.0	0.74	1.0	1.0	0.61	1.0	0.46	0.75
	DT50 (day)	5.80	1.20	6.50	0.41	231	3.92	52.8	77.0
	DT90 (day)	19.2	3.98	24.1	2.95	768	14.1	176	256
FODED	k_1 (day^{-1})	0.009	0.578	0.130	0.054	0.079	0.012	0.001	0.046
	k_2 (day^{-1})	0.162	0.578	0.007	0.890	0.000	0.192	0.056	0.001
	C_{sorb} (µg kg^{-1})	356.5	408	45.4	343	392	569	92	255
	C_{sol} (µg kg^{-1})	42	84	597	22.3	127	26	55	225
	MSE	139	448	394	2.14	677	204	846	1663
	R^2	0.984	0.854	0.985	0.998	0.668	0.989	0.484	0.782
	Curve slope	0.98	0.74	0.99	1.0	0.75	0.89	0.49	0.78
	DT50 (day)	4.98	1.20	5.89	0.85	>1000	3.83	27.3	82.2
	DT90 (day)	24.9	3.98	24.1	3.32	>1000	14.2	>1000	>1000
FOTC	k_1 (day^{-1})	0.146	0.530	0.110	0.839	0.003	0.184	0.013	0.019
	k_2 (day^{-1})	0.009	0.210	0.004	0.054	0.003	0.012	0.013	0.009
	k_r (day^{-1})	0.016	0.00	0.006	0.051	0.050	0.008	0.034	0.104
	C_0 (µg kg^{-1})	398	426	602	366	450	595	100	489
	MSE	139	495	456	2.1	1050	204	1255	2675
	R^2	0.984	0.833	0.984	0.998	0.550	0.989	0.419	0.716
	Curve slope	0.98	0.81	0.95	1.0	0.59	0.99	0.36	1.0
	DT50 (day)	4.97	1.31	6.45	0.85	138	3.84	53.3	66.8
	DT90 (day)	24.7	4.34	25.1	3.32	>100	14.2	>100	>100
MHSM	k (day^{-1})	0.113	0.578	0.104	0.611	0.003	0.167	0.013	0.009
	t_b (day)	0.00	0.00	0	0.07	0	0	0	0.43
	C_0 (µg kg^{-1})	367	408	611	279	459	575	124	428
	MSE	419	480	727	51.7	896	314	955	2378
	R^2	0.970	0.984	0.979	0.983	0.548	0.987	0.419	0.690
	Slope	1.0	0.74	1.0	1.0	0.56	1.0	0.44	0.72

	DT50 (day)	6.13	1.20	6.66	1.20	231	4.15	53.3	77.4
	DT90 (day)	20.4	3.98	22.1	3.84	767	13.8	177	256
LM	a_{max}	0.113	0.578	0.104	0.611	0.003	0.175	0.013	0.009
	r	1.00	6.06	0.97	1.0	1.00	2.34	0.82	0.96
	a_0	0.113	0.578	0.104	0.611	0.003	0.009	0.013	0.009
	C_0 (μg kg^{-1})	367	408.0	612	291	463	486	124	433
	MSE	428	499.6	727	51.7	907	294	955	2388
	R^2	0.970	0.984	0.979	0.983	0.550	0.988	0.419	0.754
	Curve slope	1.0	0.74	1.0	1.0	0.59	1.0	0.44	0.69
	DT50 (day)	6.13	1.20	6.66	1.13	>1000	5.23	53.3	53.3
	DT90 (day)	20.4	3.98	22.1	3.77	>1000	14.4	177	77.0

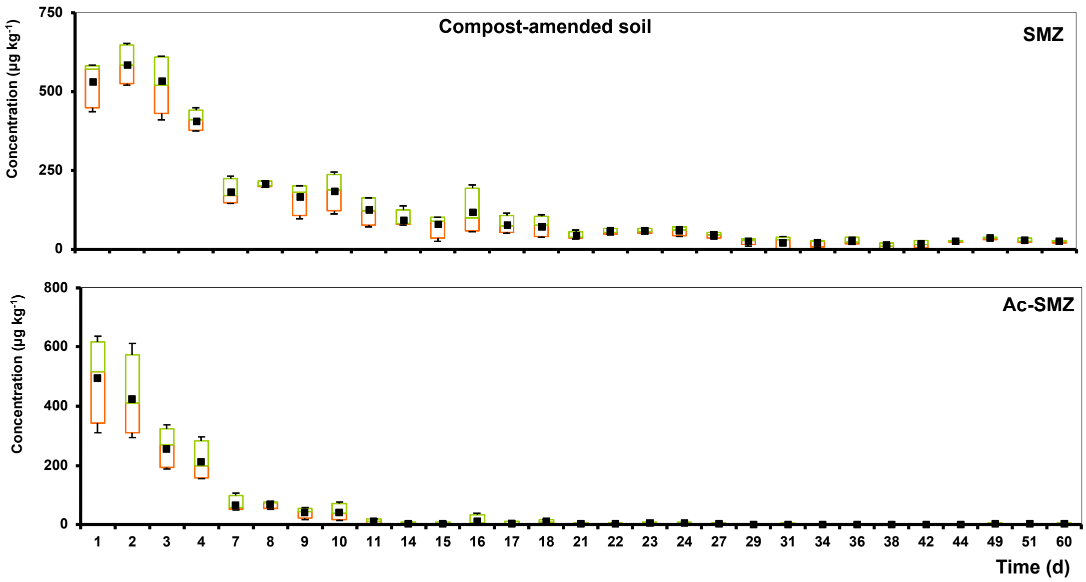
299 **Supplementary Table 6. (Continuation)**

		IBU	1-OH-IBU	2-OH-IBU	CBX-IBU	MeP	OH-MeP	PrP	3,4-DHB	4-HB
SFO	k	0.081	0.245	0.011	0.227	0.13	0.200	0.078	0.41	1.32
	C_0 (μg kg^{-1})	290	433	437	298	123	621	11.08	425	411
	MSE	321	617	6376	695	119	189	1.33	144	11.5
	R^2	0.951	0.870	0.501	0.769	0.835	0.994	0.738	0.930	0.954
	Slope	0.95	0.93	0.51	0.78	0.94	1.0	1.0	1.0	1.0
	DT50 (day)	8.56	2.83	63.0	3.05	5.33	3.47	8.89	1.69	0.53
	DT90 (day)	28.4	9.40	209	10.1	17.7	11.5	29.5	5.62	1.74
BEM	k_1 (day^{-1})	0.076	0.187	0.012	0.102	0.114	0.164	0.078	0.32	1.131
	k_2 (kg μg^{-1} day^{-1})	0	1.58x10^{-4}	0	0.0013	0.0005	4.82x10^{-5}	0	9.91x10^{-7}	0.003
	C_0 (μg kg^{-1})	276	423	456	471	149	575	11.1	334	737
	MSE	338	599	6503	629	107	276	1.33	222	10.5
	R^2	0.951	0.871	0.495	0.796	0.879	0.987	0.738	0.889	0.956
	Slope	0.91	0.87	0.51	0.80	1.0	0.97	1.0	0.84	1.0
	DT50 (day)	9.12	2.95	57.8	1.31	4.15	3.77	8.89	2.16	0.26
	DT90 (day)	30.3	10.9	192	8.10	16.3	13.2	29.5	7.19	1.24
FODED	k_1 (day^{-1})	0.081	0.149	0.011	0.213	0.310	0.231	0.078	0.33	1.32
	k_2 (day^{-1})	0.180	0.711	0.011	0.218	0.094	0.013	0.078	2.78	1.32
	C_{sorb} (μg kg^{-1})	0.0	327	260	288.2	96.5	25.7	9.16	318	206
	C_{sol} (μg kg^{-1})	290	225	177	0	33.9	620	1.92	1043	206
	MSE	321	393	6376	698	103	71.4	1.33	113	11.48
	R^2	0.951	0.922	0.501	0.768	0.847	0.996	0.738	0.970	0.954
	Slope	0.95	0.90	0.51	0.76	0.93	0.99	1.0	1.0	1.0
	DT50 (day)	8.56	1.66	63.01	3.18	5.30	3.17	8.89	1.33	0.53
	DT90 (day)	28.4	9.48	>1000	10.6	21.3	11.6	29.5	6.17	1.74
FOTC	k_1 (day^{-1})	0.081	0.236	0.011	0.241	0.122	0.157	0.078	1.89	1.32
	k_2 (day^{-1})	0.081	0.236	0.011	0.153	0.057	0.157	0.078	0.325	1.32
	k_r (day^{-1})	0.097	3.061	0.199	0.045	0.060	0.110	0.123	0.610	0.033
	C_0 (μg kg^{-1})	290	424	437	302	104	547	11.1	1123	411
	MSE	321	171	6376	691	127	461	1.33	113	11.5
	R^2	0.951	0.968	0.501	0.771	0.834	0.975	0.738	0.970	0.954
	Slope	0.95	0.98	0.52	0.76	0.72	0.98	1.0	1.1	1.0
	DT50 (day)	8.56	2.94	63.0	2.95	6.31	4.41	8.89	0.42	0.53
	DT90 (day)	28.4	9.76	209	10.5	28.1	14.7	29.5	3.18	1.74
MHSM	k (day^{-1})	0.081	0.245	0.011	0.227	0.125	0.200	0.078	0.409	1.32
	t_b (day)	0	0	0.00	0	0	0	0	3.98x10^{-8}	1.36
	C_0 (μg kg^{-1})	290	433	437	298	123	621	11.1	425	68.4
	MSE	321	617	6376	695	119	189	1.33	144	11.5
	R^2	0.951	0.870	0.501	0.769	0.835	0.994	0.738	0.930	0.954
	Slope	0.95	0.92	0.52	0.78	0.94	1.0	1.0	1.0	1.0
	DT50 (day)	8.56	2.83	63.0	3.05	5.55	3.47	8.89	1.69	1.89
	DT90 (day)	28.4	9.42	209	10.1	18.4	11.5	29.5	5.63	3.10
LM	a_{max}	0.081	0.245	0.017	0.227	0.13	0.200	0.09	0.409	1.32

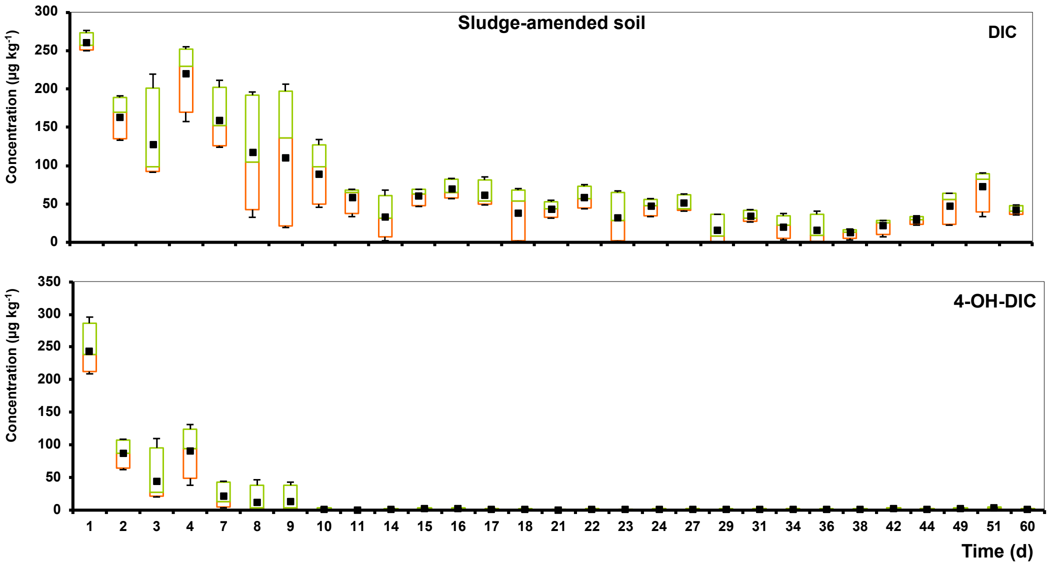
r	1.00	1.00	0.450	1.01	1.00	0.99	1.00	0.72	1.0
a_0	0.081	0.245	9.36×10^{-7}	0.227	0.13	0.200	0.09	0.409	1.32
C_0 (μg kg^{-1})	290	433	400	298	151	621	11.5	425	639
MSE	321	617	4507	695	196	189	1.36	144	164
R^2	0.951	0.870	0.634	0.769	0.834	0.994	0.742	0.930	0.956
Slope	0.95	0.93	0.68	0.78	1.2	1.0	1.2	1.0	1.6
DT50 (day)	8.56	2.83	62.6	3.05	5.33	3.47	7.70	1.69	0.53
DT90 (day)	28.4	9.40	157.2	10.1	17.7	11.5	25.6	5.63	1.74



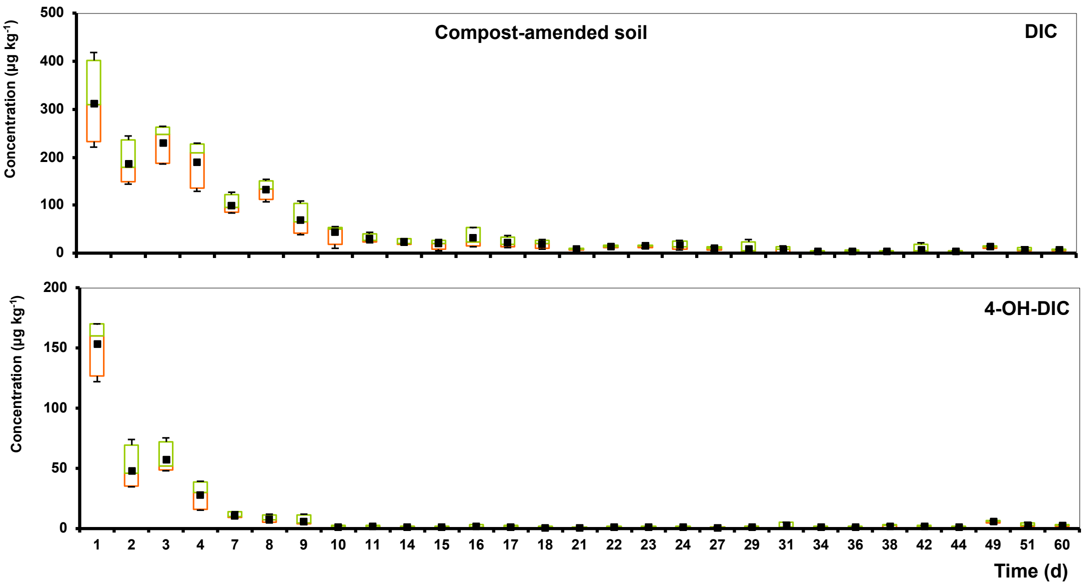
Supplementary Figure 1. Concentrations of SMZ and AcSMZ in sludge-amended soil.



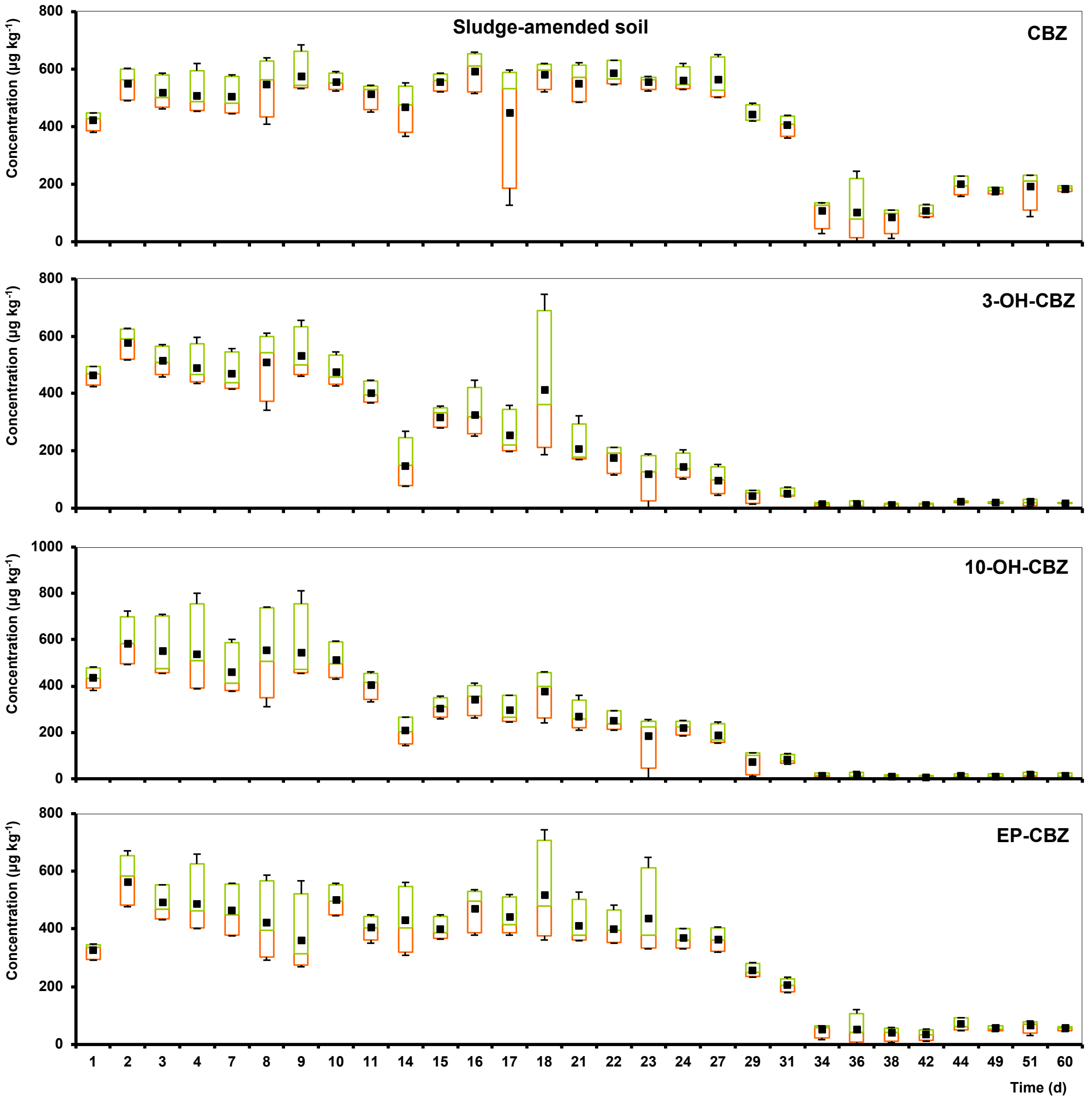
Supplementary Figure 2. Concentrations of SMZ and AcSMZ in compost-amended soil.



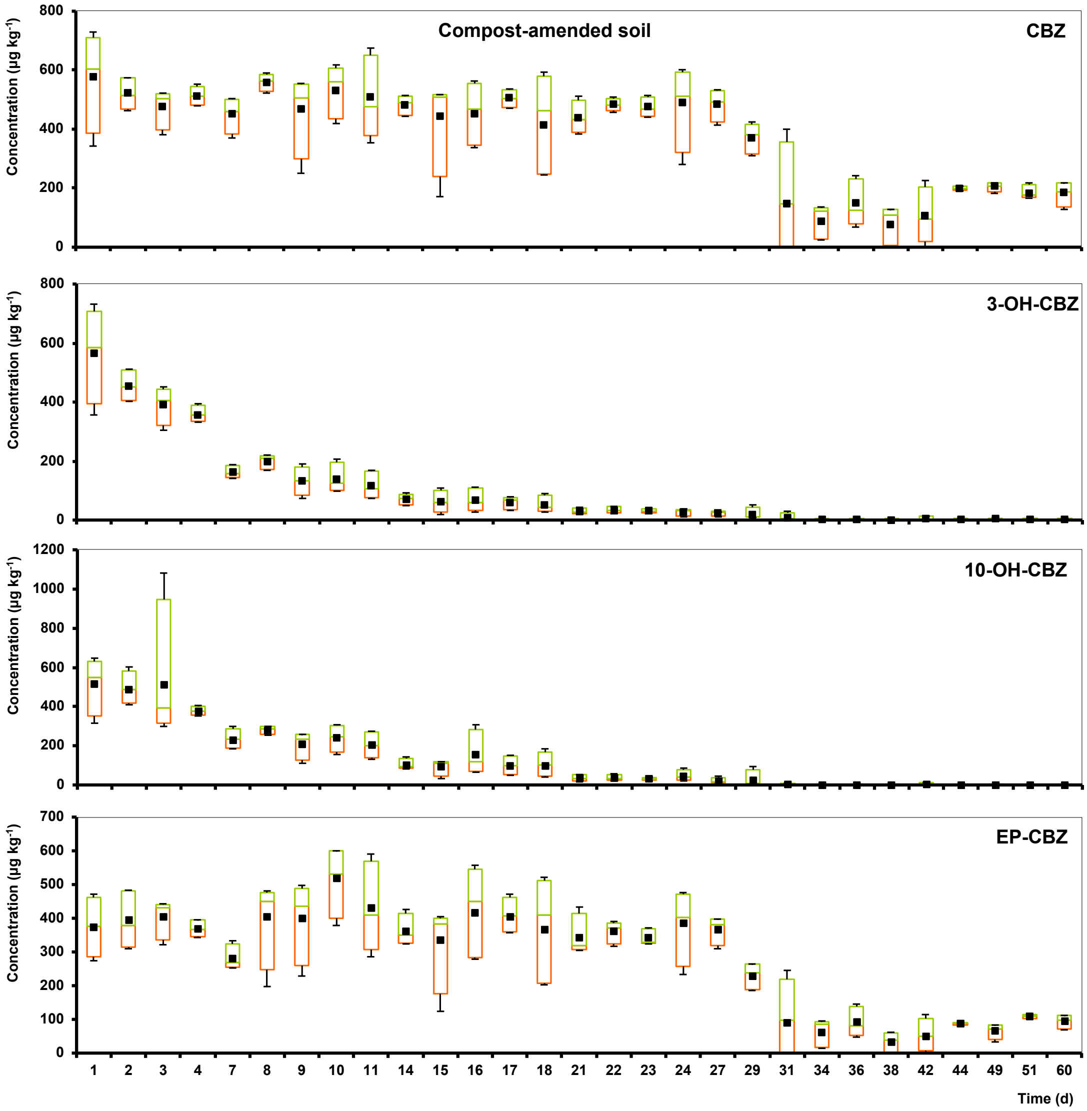
Supplementary Figure 3. Concentrations of DIC and 4-OH-DIC in sludge-amended soil



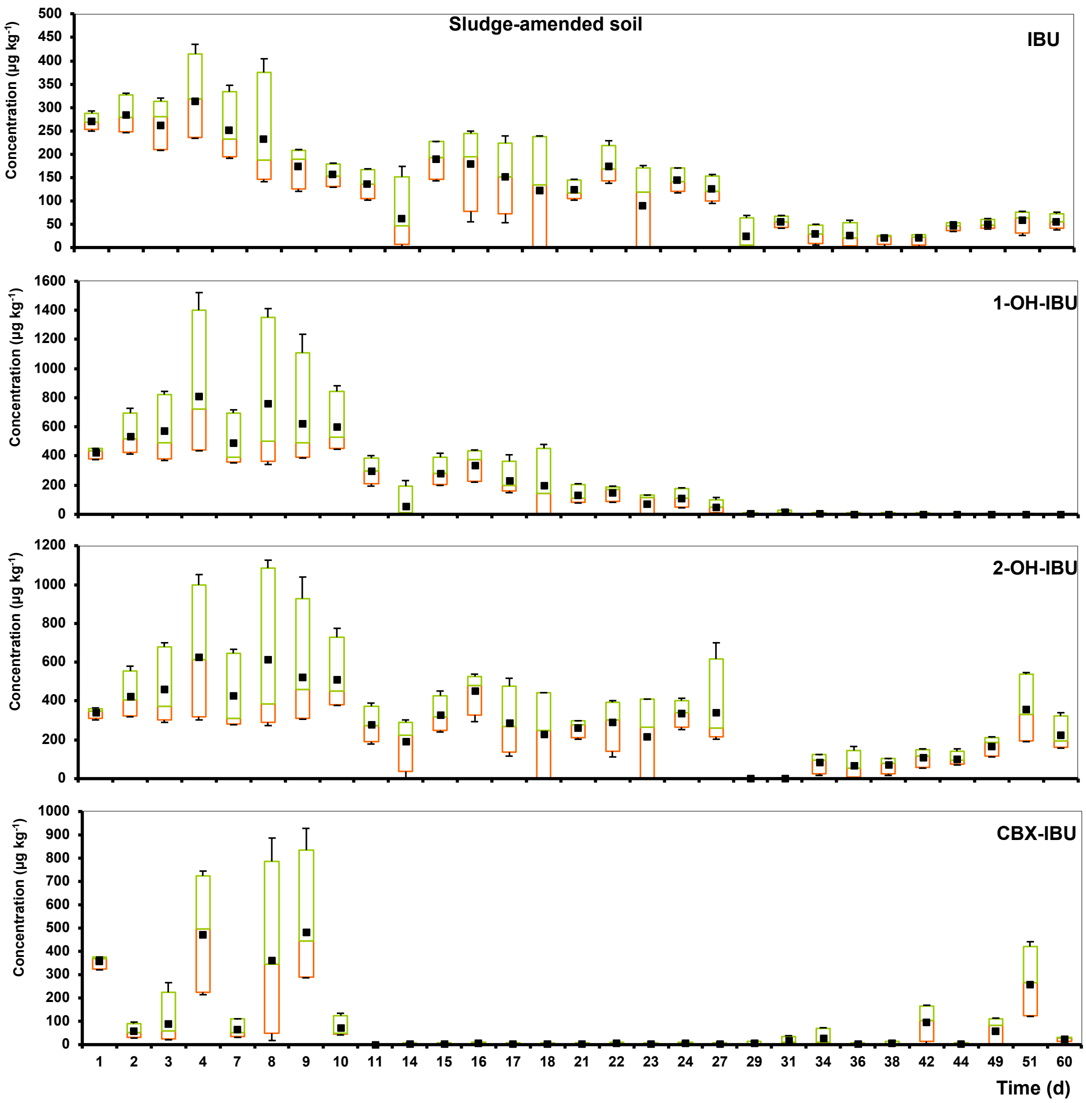
Supplementary Figure 4. Concentrations of DIC and 4-OH-DIC measured in compost-amended soil



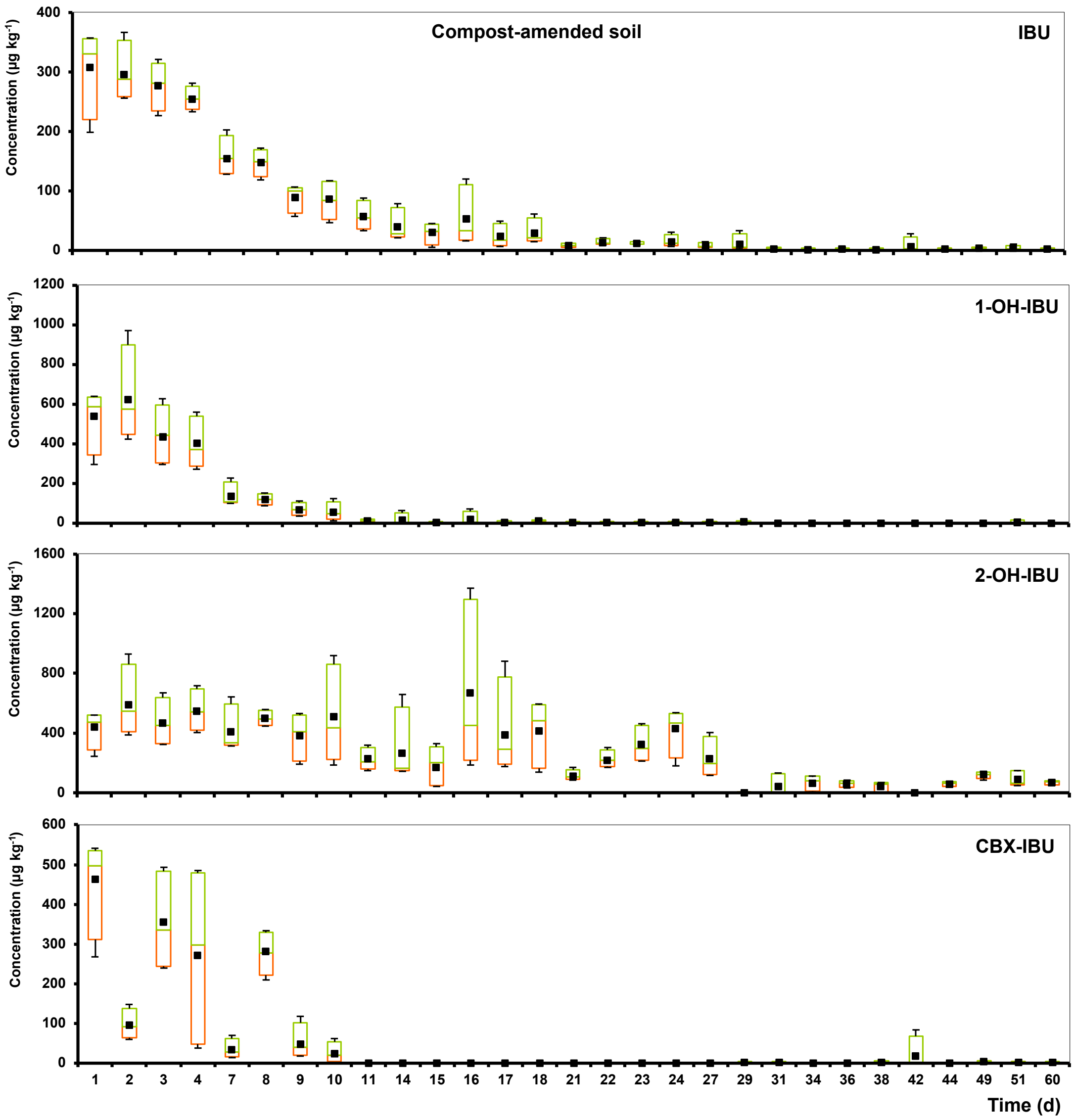
Supplementary Figure 5. Concentrations of CBZ and its metabolites in sludge-amended soil



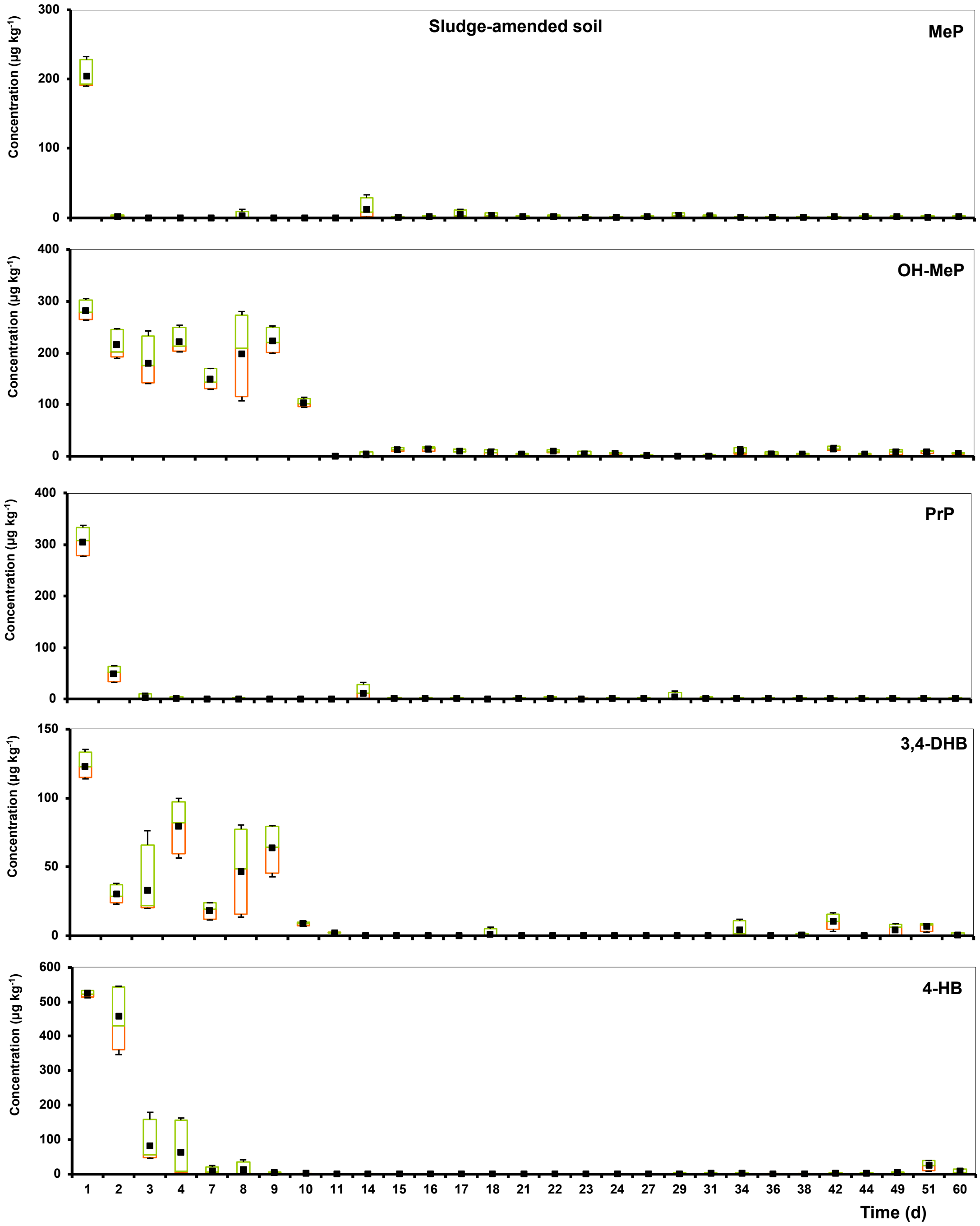
Supplementary Figure 6. Concentrations of CBZ and its metabolites in compost-amended soil



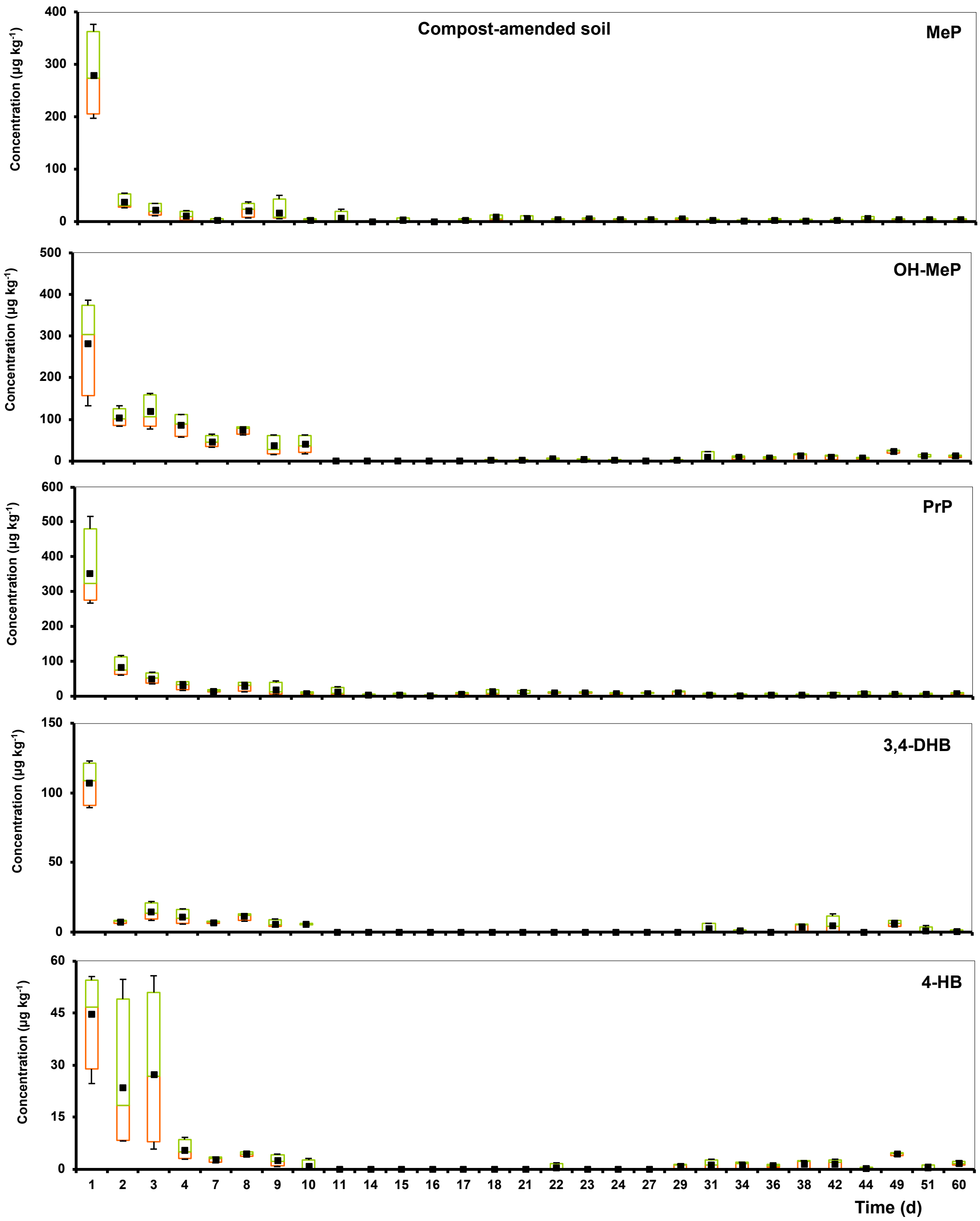
Supplementary Figure 7. Concentrations of IBU and its metabolites in sludge-amended soil



Supplementary Figure 8. Concentrations of IBU and its metabolites in compost-amended soil



Supplementary Figure 9. Concentrations of parabens and their metabolites in sludge-amended soil



Supplementary Figure 10. Concentrations of parabens and their metabolites in compost-amended soil**.**