Supplementary material for:

Environmental risk assessment of perfluoroalkyl substances and halogenated flame retardants released from biosolids-amended soils

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S.1. Material and Methods: Study design

S1.1. Experimental details to study the transfer and fate of the selected emerging compounds to different environmental compartments

Four different organic wastes were selected for the present study: an aerobically digested municipal solid waste (MSW) compost (B-1), an anaerobically digested thermal drying sludge (B-2), an aerobically digested composted sewage sludge (B-3) and an anaerobically digested MSW compost (B-4). In previous works, these biosolids were applied to soil in different experiments to study the transfer and fate of the selected emerging compounds to different environmental compartments. Firstly, the biosolids were characterized to determine PFAS and HFR concentrations and then were used to study the transfer and bioaccumulation of selected emerging organic compounds from biosolid-amended soil to earthworm (*Eisenia andrei*) using MS·3 systems (Navarro et al., 2016). In another experiment, we conducted semi-field tests simulating natural conditions to determine the leaching and runoff capacity of PFASs and HFRs following the fortification and application of municipal biosolids (Navarro et al., 2018). The four biosolid selected (B-1, B-2, B-3 and B-4) were fortified with perfluorooctanesulfonate (PFOS; ~1 mg PFOS per kg biosolid), decabromodiphenyl ether (c-decaBDE; ~10 mg/kg) and Dechlorane Plus (DP;~0.26 mg/kg) commercial mixtures and were applied to soil packed in 15 runoff-leaching trays (2.5 x 2 x 0.05 m). These trays were designed to collect the leachate and runoff water generated by natural rainfall. In another experiment, the bioaccumulation behaviour of PFASs and HFRs was examined in tomato plant (*Solanum lycopersicum L.*), which was grown in field soil amended with a single application of biosolids B-2 and B-4, respectively, at agronomic rate for nitrogen (Navarro et al., 2017). The concentrations measured for perfluorooctanesulfonate (PFOS), perfluorooctanoic acid (PFOA), pentabrominated diphenyl ether (penta-BDE: sum of BDE-85, -99 and -100), decabrominated diphenyl ether (Deca-BDE: BDE-209), Declorane Plus (DP: sum of *anti*- and *syn*-DP isomers) and decabromodiphenyl ethane (DBDPE) in the different environmental compartments (biosolids, runoff water and tomato fruit) were used in the exposure assessment (Table 1 and Table S1).

S1.2. Estimation of the biosolid application rates

To calculate the application rates, it was necessary to analyze nitrogen content. To determine the required amount of biosolid to be added to soil and guarantee agronomic conditions, an equivalent of 150 kg of the available nitrogen form ($N_{available}$)/ha was considered appropriate; $N_{available}$ was calculated following the EPA (1995) recommendations. The $N_{available}$ to plants during the growing season was estimated by the following equation:

$$N_{available} = N - NO_3^- + (1 - \alpha)\, N - NH_4^+ + \beta N_{org}$$

Eq. (S1)

where α is the percentage of $N\text{-}NH_4^+$ volatilized. When mixing the dry residue with dry soil, the volatilization of ammonia was estimated to be minor, ≈ 10%, α = 0.1, and β was the percentage of mineralised N_{org}. When sludge is thermally dried, its organic matter is very stable and the mineralisation process of the N that these residues have is slow, as only 10% is available during the culture period ≈ 10%, β = 0.1.

Table S1. Concentrations detailed of the compounds studied in each different environmental compartment.

	B-1	**B-2**	**B-3**	**B-4**
Biosolid (ng/g)[a]				
PFOS	14.6	28.2	64.4	0.65
PFOA	3.65	8.90	9.35	2.52
Penta-BDE	1.82	15.2	17.3	9.60
Deca-BDE	29.3	228	347	130
DP	2.28	14.6	12.1	5.50
DBDPE	N.D.	0.13	0.10	0.15
Runoff (ng/L)[b,c]				
PFOS	326	219	268	819
PFOA	23.4	33.2	16.8	17.1

Penta-BDE	0.46	0.34	0.45	0.37
Deca-BDE	200	92.4	274	364
DP	14.4	16.6	13.6	29.8
DBDPE	0.06	0.43	2.48	5.58
Tomato fruit (ng/g w.w.)[d]				
PFOS	N.A.	0.003	N.A.	0
PFOA	N.A.	0.014	N.A.	0
Penta-BDE	N.A.	0.007	N.A.	0.01
Deca-BDE	N.A.	0.671	N.A.	0.671
DP	N.A.	0.011	N.A.	0.007
DBDPE	N.A.	N.D.	N.A.	N.D.

N.D.: not detected. N.A.: not available.

B-1: aerobic-digested MSW compost, B-2: anaerobic-digested thermal drying sludge,

B-3: aerobic-digested composted sewage sludge, B-4: anaerobic-digested MSW compost.

Penta-BDE: sum of BDE-85, -99 and -100.

[a]Navarro et al., 2016. [b]Navarro et al., 2018.

[c]Concentrations of Penta-BDE, Deca-BDE, DP and DBDPE have been normalized to ng/L.

[d]Navarro et al., 2017. Only B-2 and B-4 were used in this experiment.

Table S2. Determination of nitrogen in the biosolids to calculate the application rates in agronomic conditions.

Parameter		**B-1**	**B-2**	**B-3**	**B-4**
$\mathbf{N_{org}}$	g/kg	13.5	55.4	25.3	10.3
$\mathbf{N\text{-}NH_4^+}$	g/kg	0.83	3.03	10.1	1.2
$\mathbf{N\text{-}NO_3^-}$	g/kg	0.77	0.16	0.25	0.44
$\mathbf{N_{available}}$	g/kg	2.87	8.42	11.9	2.56
$\mathbf{APPL_{waste}}$	kg/m^2·año	5.20	1.80	1.20	5.90

$APPL_{waste}$: waste application rate

Table S3. General parameters used in the environmental exposure assessment for the different compartments.

Parameters			
$DEPTH_{soil}$	Mixing depth of soil	m	0.20[a]
RHO_{water}	Density of the water phase	kg/m^3	1000
RHO_{air}	Density of air	kg/m3	1.3
RHO_{solid}	Density of the solid phase	kg/m^3	2500
RHO_{soil}	Bulk density of soil	kg/m^3	1700
RHO_{susp}	Bulk density of suspended matter	kg/m^4	9250
$RHO_{earthworm}$	Density of earthworm	kg/L	1
$Fwater_{soil}$	Fraction water in compartment soil	m^3/m^3	0.2
$Fwater_{susp}$	Fraction water in suspended matter	m^3/m^3	9
$Fair_{soil}$	Fraction air in compartment soil	m^3/m^3	0.2
$Fair_{susp}$	Fraction air in suspended matter (only relevant for soil)	m^3/m^3	0.2
$Fsolid_{soil}$	Fraction solids in compartment soil	m^3/m^3	0.6
$Fsolid_{susp}$	Fraction solids in suspended matter	m^3/m^3	0.1
Foc_{soil}	Weight fraction organic carbon in soil solids	kg/kg	0.01[b]
Foc_{susp}	Weight fraction of organic carbon in suspended solids	kg/kg	0.1
F_{gut}	Fraction of gut loading in worm	kg/kg	0.1
$kasl_{air}$	Partial mass transfer coeff. at air-side of the air-soil interface	m/d	120
$kasl_{soil}$	Partial mass transfer coeff. at soil air-side of the air-soil interface	m/d	0.48
$Finf_{soil}$	Fraction of rain water that infiltrates into soil	---	0.25
$CONV_{soil}$	Conversion factor for soil concentration wet-dry weight soil	kg/kg	1.13

$SUSP_{water}$	Concentration of suspended matter in the river	mg/L	15
RAIN	Rate of wet precipitation	m/d	0.00177[c]
DILUTION	Dilution factor	---	10
R	Gas constant	Pa m^3/ mol k	8.314
TEMP	Temperature at the air-water interface	K	285
T	Time	d	365

Data obtained by ECHA' recommendations (ECHA, 2016).

[a]The depth of soil represents the depth range for the top soil layer which is of interest. The depth of 0.20 m is taken because this range represents the ploughing depth (ECHA, 2016).

[b]This value (0.01 kg/kg) was applied instead of 0.02 kg/kg (ECHA, 2016) due to the poor organic matter content in Spanish soils.

[c]Value calculated considering the average annual rainfall in Spain (649 mm/year).

Table S4. Specific parameters used in the environmental exposure assessment for the different compartments.

Parameters		PFOS	PFOA	Penta-BDE	Deca-BDE	DP	DBDPE
Log K_{ow}	---	4.49[a]	4.81[a]	7.66[a]	12.11[a]	11.27[a]	13.64[a]
Log K_{oc}	L/kg	3.41[a]	2.82[a]	4.76[a]	7.68[a]	7.68[a]	6.38[a]
HENRY	Pa m^3/mol	3.19×10^{-4}[b]	2.533[c]	0.121[a]	0.0012[a]	0.754[a]	0.00651[a]
BIO	---	NO[a]	NO[a]	NO[a]	NO[a]	NO[a]	NO[a]
$DT50bio_{soil}$	d	300	300	4650[d]	10230[d]	30000	30000
$BCF_{earthworm}$	L/kg	372	776	5.49×10^{5}	1.55×10^{10}	2.23×10^{9}	5.24×10^{11}
BCF_{fish}	L/kg	3.16[a]	3.16[a]	6320[a]	41.71[a]	107[a]	7.43[a]
BMF	---	2	2	10	1	1	10
$K_{air\text{-}water}$	m^3/m^3	1.34×10^{-7}	0.0011	5.11×10^{-5}	5.06×10^{-7}	3.18×10^{-4}	2.75×10^{-6}
$K_{soil\text{-}water}$	m^3/m^3	39	10	871	716294	721259	36066
$K_{susp\text{-}water}$	m^3/m^3	73	25	1461	1193832	1202107	60118
Kp_{soil}	L/kg	26	7	581	477529	480839	24044
Kp_{susp}	L/kg	256	65	5808	4775293	4808393	240436
k_{volat}	1/d	2.09×10^{-6}	0.0623	3.52×10^{-5}	4.24×10^{-10}	2.65×10^{-7}	4.57×10^{-8}
k_{leach}	1/d	5.72×10^{-5}	2.21×10^{-4}	2.54×10^{-6}	3.09×10^{-9}	3.07×10^{-9}	6.13×10^{-8}
$kbio_{soil}$	1/d	0.0023	0.0023	1.49×10^{-4}	6.78×10^{-5}	2.31×10^{-5}	2.31×10^{-5}
k	1/d	0.0024	0.065	1.87×10^{-4}	6.78×10^{-5}	2.34×10^{-5}	2.32×10^{-5}
Facc	---	0.0042	5.21×10^{-11}	0.934	0.976	0.992	0.992

Log K_{ow}: octanol-water partitioning coefficient; Log K_{oc}: partition coefficient organic carbon-water; HENRY: Henry's law constant;

BIO: tendency to biodegradation; $DT50bio_{soil}$: half-life for biodegradation in bulk soil; $BCF_{earthworm}$: bioconcentration factor for earthworms on wet weight;

BCF_{fish}: bioconcentration factor (fish); BMF: biomagnification factor in fish. $K_{air\text{-}water}$: air-water partitioning coefiicient; $K_{soil\text{-}water}$: soil-water partitioning coefficient;

$Kp_{susp\text{-}water}$: suspended matter-water partitioning coefficient; Kp_{soil}: partition coefficient solid-water in soil; Kp_{susp}: partition coefficient solid-water in suspended matter.

k_{volat}: pseudo-first order rate constant for volatilisation from soil; k_{leach}:pseudo-first order rate constant for leaching from top soil;

$kbio_{soil}$: pseudo-first order rate constant for biodegradation in soil; k: first order rate constant for removal from top soil; Facc: fraction accumulation in one year;

Data obtained by ECHA' recommendations (ECHA, 2016).

[a]Data obtained by EPISuite 4.1. [b]Brooke et al., 2004. [c]Stemmler and Lammel, 2010. [d]Andrade et al., 2010.

Table S5. Predicted environmental concentrations in the different environmental compartments: PEC_{soil} (mg/kg), $PEC_{oral,predator (T)}$ (mg/kg), PEC_{water} (mg/L), PEC_{sed} (mg/kg), $PEC_{oral, predator (Aq)}$ (mg/kg).

	B-1	**B-2**	**B-3**	**B-4**
PFOS				
PEC_{soil}	1.49×10^{-4}	9.99×10^{-5}	1.52×10^{-4}	7.55×10^{-6}
$PEC_{oral, predator (T)}$	2.21×10^{-3}	1.48×10^{-3}	2.25×10^{-3}	1.12×10^{-4}
PEC_{water}	3.25×10^{-5}	2.18×10^{-5}	2.67×10^{-5}	8.15×10^{-5}
PEC_{sed}	2.57×10^{-4}	1.72×10^{-4}	2.11×10^{-4}	6.45×10^{-4}
$PEC_{oral, predator (Aq)}$	2.05×10^{-4}	1.38×10^{-4}	1.69×10^{-4}	5.16×10^{-4}
PFOA				
PEC_{soil}	2.36×10^{-6}	1.99×10^{-6}	1.39×10^{-6}	1.85×10^{-6}
$PEC_{oral, predator (T)}$	2.79×10^{-4}	2.35×10^{-4}	1.65×10^{-4}	2.18×10^{-4}
PEC_{water}	2.34×10^{-6}	3.32×10^{-6}	1.68×10^{-6}	1.71×10^{-6}
PEC_{sed}	6.42×10^{-6}	9.10×10^{-6}	4.61×10^{-6}	4.69×10^{-6}
$PEC_{oral, predator (Aq)}$	1.48×10^{-5}	2.10×10^{-5}	1.06×10^{-5}	1.08×10^{-5}
Penta-BDE				
PEC_{soil}	2.69×10^{-5}	7.78×10^{-5}	5.90×10^{-5}	1.61×10^{-4}
$PEC_{oral, predator (T)}$	0.026	0.075	0.057	0.155
PEC_{water}	4.19×10^{-8}	3.14×10^{-8}	4.12×10^{-8}	3.40×10^{-8}
PEC_{sed}	6.61×10^{-6}	4.96×10^{-6}	6.51×10^{-6}	5.37×10^{-6}
$PEC_{oral, predator (Aq)}$	2.65×10^{-3}	1.99×10^{-3}	2.60×10^{-3}	2.15×10^{-3}
Deca-BDE				
PEC_{soil}	4.42×10^{-4}	1.19×10^{-3}	1.21×10^{-3}	2.23×10^{-3}
$PEC_{oral, predator (T)}$	14.6	39.3	39.9	73.4
PEC_{water}	2.75×10^{-7}	1.27×10^{-7}	3.77×10^{-7}	5.01×10^{-7}
PEC_{sed}	0.036	0.016	0.049	0.065
$PEC_{oral, predator (Aq)}$	1.15×10^{-5}	5.31×10^{-6}	1.57×10^{-5}	2.09×10^{-5}
DP				
PEC_{soil}	3.47×10^{-5}	7.70×10^{-5}	4.25×10^{-5}	9.50×10^{-5}
$PEC_{oral, predator (T)}$	0.164	0.364	0.201	0.450
PEC_{water}	1.97×10^{-8}	2.26×10^{-8}	1.86×10^{-8}	4.07×10^{-8}
PEC_{sed}	2.56×10^{-3}	2.94×10^{-3}	2.42×10^{-3}	5.29×10^{-3}
$PEC_{oral, predator (Aq)}$	2.12×10^{-6}	2.43×10^{-6}	2.00×10^{-6}	4.38×10^{-6}
DBDPE				
PEC_{soil}	---	6.63×10^{-7}	3.35×10^{-7}	2.58×10^{-6}
$PEC_{oral, predator (T)}$	---	14.7	74.3	57.3
PEC_{water}	1.24×10^{-9}	9.29×10^{-9}	5.39×10^{-8}	1.21×10^{-7}

PEC_{sed}	8.09×10^{-6}	6.04×10^{-5}	3.50×10^{-4}	7.87×10^{-4}
$PEC_{oral, predator (Aq)}$	9.24×10^{-8}	6.90×10^{-7}	4.01×10^{-6}	8.99×10^{-6}

PEC_{soil}: Predicted environmental concentration in soil (mg/kg).

$PEC_{oral,predator (T)}$: Predicted environmental concentration of contaminant in the food (earthworm) of worm-eating birds or mammals (mg/kg $_{wet\ earthworm}$).

PEC_{water}: Predicted environmental concentration in surface water during release episode (mg/L).

PEC_{sed}: Predicted environmental concentration in sediment (mg/kg)

$PEC_{oral,predator (Aq)}$: Predicted environmental concentration of contaminant in the food (fish) of fish-eating predators (mg/kg $_{wet\ fish}$).

Table S6. Predicted no effect concentrations in the differents environmental compartments: $PNEC_{soil}$ (mg/kg), $PNEC_{oral}$ (mg/kg), $PNEC_{water}$ (mg/L), $PNEC_{sed}$ (mg/kg).

	$PNEC_{soil}$ (mg/kg)	**$PNEC_{oral}$** (mg/kg)	**$PNEC_{water}$** (µg/L)	**$PNEC_{sed}$** (mg/kg)
PFOS	0.1[a]	0.0167[b]	25[b]	0.067[c]
PFOA	0.16[a]	0.158[d]	20[d]	0.143[e]
Penta-BDE	0.38[f]	1[g]	0.53[g]	0.31[g]
Deca-BDE	98[f]	2500[h]	0.2[c]	127[c]
DP	0.075[i]	1179.8[i]	---	---
DBDPE	156.3[j]	220[k]	0.2[c]	100[l]

$PNEC_{soil}$: Predicted no effect concentration in soil.

$PNEC_{oral}$: Predicted no effect concentration for secondary poisoning of birds and mammals.

$PNEC_{water}$: Predicted no effect concentration in water.

$PNEC_{sed}$: Predicted no effect concentration in sediment.

[a]SFT, 2008. [b]Brooke et al., 2004. [c]Møskeland, 2010. [d]Environment Canada, 2012.

[e]Value calculated from $PNEC_{water}$. [f]Cincinelli et al., 2012. [g]European Chemicals Bureau, 2001.

[h]European Chemicals Bureau, 2002. [i]Data based on analogue chlordane and mirex; ECCC, 2016.

[j]Hardy et al., 2011. [k]Dungey and Akintoye, 2007. [l]Hardy et al., 2012.

Table S7. Predicted environmental concentrations in soil obtained considering the biosolid annual application.

	B-1	**B-2**	**B-3**	**B-4**
PFOS				

$PEC_{porewater}$	mg/L	6.57×10^{-6}	4.39×10^{-6}	6.69×10^{-6}	3.32×10^{-7}
C_{soil} (0)	mg/kg	2.23×10^{-4}	1.49×10^{-4}	2.27×10^{-4}	1.13×10^{-5}
PEC_{soil} (1 year)	mg/kg	1.49×10^{-4}	9.99×10^{-5}	1.52×10^{-4}	7.55×10^{-6}
PEC_{soil} (10 years)	mg/kg	3.86×10^{-4}	2.58×10^{-4}	3.92×10^{-4}	1.95×10^{-5}
PEC_{soil} (25 years)	mg/kg	3.86×10^{-4}	2.58×10^{-4}	3.93×10^{-4}	1.95×10^{-5}
PEC_{soil} (50 years)	mg/kg	3.86×10^{-4}	2.58×10^{-4}	3.93×10^{-4}	1.95×10^{-5}
PEC_{soil} (100 years)	mg/kg	3.86×10^{-4}	2.58×10^{-4}	3.93×10^{-4}	1.95×10^{-5}
PFOA					
$PEC_{porewater}$	mg/L	4.00×10^{-7}	3.38×10^{-7}	2.36×10^{-7}	3.13×10^{-7}
C_{soil} (0)	mg/kg	5.58×10^{-5}	4.71×10^{-5}	3.30×10^{-5}	4.37×10^{-5}
PEC_{soil} (1 year)	mg/kg	2.36×10^{-6}	1.99×10^{-6}	1.39×10^{-6}	1.85×10^{-6}
PEC_{soil} (10 years)	mg/kg	5.58×10^{-5}	4.71×10^{-5}	3.30×10^{-5}	4.37×10^{-5}
PEC_{soil} (25 years)	mg/kg	5.58×10^{-5}	4.71×10^{-5}	3.30×10^{-5}	4.37×10^{-5}
PEC_{soil} (50 years)	mg/kg	5.58×10^{-5}	4.71×10^{-5}	3.30×10^{-5}	4.37×10^{-5}
PEC_{soil} (100 years)	mg/kg	5.58×10^{-5}	4.71×10^{-5}	3.30×10^{-5}	4.37×10^{-5}
Penta-BDE					
$PEC_{porewater}$	mg/L	5.25×10^{-8}	1.52×10^{-7}	1.15×10^{-7}	3.14×10^{-7}
C_{soil} (0)	mg/kg	2.78×10^{-5}	8.05×10^{-5}	6.10×10^{-5}	1.67×10^{-4}
PEC_{soil} (1 year)	mg/kg	2.69×10^{-5}	7.78×10^{-5}	5.90×10^{-5}	1.61×10^{-4}
PEC_{soil} (10 years)	mg/kg	2.09×10^{-4}	6.03×10^{-4}	4.58×10^{-4}	1.25×10^{-3}
PEC_{soil} (25 years)	mg/kg	3.46×10^{-4}	9.99×10^{-4}	7.58×10^{-4}	2.07×10^{-3}
PEC_{soil} (50 years)	mg/kg	4.08×10^{-4}	1.18×10^{-3}	8.96×10^{-4}	2.44×10^{-3}
PEC_{soil} (100 years)	mg/kg	4.22×10^{-4}	1.22×10^{-3}	9.25×10^{-4}	2.52×10^{-3}
Deca-BDE					
$PEC_{porewater}$	mg/L	1.05×10^{-9}	2.83×10^{-9}	2.87×10^{-9}	5.29×10^{-9}
C_{soil} (0)	mg/kg	4.48×10^{-4}	0.001	0.001	0.002
PEC_{soil} (1 year)	mg/kg	4.42×10^{-4}	0.001	0.001	0.002
PEC_{soil} (10 years)	mg/kg	0.004	0.011	0.011	0.020
PEC_{soil} (25 years)	mg/kg	0.009	0.023	0.023	0.043
PEC_{soil} (50 years)	mg/kg	0.013	0.035	0.036	0.066
PEC_{soil} (100 years)	mg/kg	0.017	0.045	0.046	0.085
PEC_{soil} (250 years)	mg/kg	0.018	0.049	0.050	0.092
PEC_{soil} (500 years)	mg/kg	0.018	0.049	0.050	0.092
DP					
$PEC_{porewater}$	mg/L	8.18×10^{-11}	1.81×10^{-10}	1.00×10^{-10}	2.24×10^{-10}
C_{soil} (0)	mg/kg	3.49×10^{-5}	7.73×10^{-5}	4.27×10^{-5}	9.54×10^{-5}

PEC_{soil} (1 year)	mg/kg	3.47×10^{-5}	7.70×10^{-5}	4.25×10^{-5}	9.50×10^{-5}
PEC_{soil} (10 years)	mg/kg	3.36×10^{-4}	7.44×10^{-4}	4.11×10^{-4}	9.19×10^{-4}
PEC_{soil} (25 years)	mg/kg	7.88×10^{-4}	0.002	9.65×10^{-4}	0.002
PEC_{soil} (50 years)	mg/kg	0.001	0.003	0.002	0.004
PEC_{soil} (100 years)	mg/kg	0.002	0.005	0.003	0.006
PEC_{soil} (250 years)	mg/kg	0.004	0.008	0.004	0.010
PEC_{soil} (500 years)	mg/kg	0.004	0.009	0.005	0.011
PEC_{soil} (750 years)	mg/kg	0.004	0.009	0.005	0.011
DBDPE					
$PEC_{porewater}$	mg/L	---	3.13×10^{-11}	1.58×10^{-11}	1.22×10^{-10}
C_{soil} (0)	mg/kg	---	6.66×10^{-7}	3.36×10^{-7}	2.59×10^{-6}
PEC_{soil} (1 year)	mg/kg	---	6.63×10^{-7}	3.35×10^{-7}	2.58×10^{-6}
PEC_{soil} (10 years)	mg/kg	---	6.41×10^{-6}	3.24×10^{-6}	2.50×10^{-5}
PEC_{soil} (25 years)	mg/kg	---	1.51×10^{-5}	7.61×10^{-6}	5.87×10^{-5}
PEC_{soil} (50 years)	mg/kg	---	2.73×10^{-5}	1.38×10^{-5}	1.06×10^{-4}
PEC_{soil} (100 years)	mg/kg	---	4.51×10^{-5}	2.28×10^{-5}	1.76×10^{-4}
PEC_{soil} (250 years)	mg/kg	---	6.95×10^{-5}	3.51×10^{-5}	2.68×10^{-4}
PEC_{soil} (500 years)	mg/kg	---	7.78×10^{-5}	3.93×10^{-5}	3.03×10^{-4}
PEC_{soil} (750 years)	mg/kg	---	7.88×10^{-5}	3.98×10^{-5}	3.07×10^{-4}
PEC_{soil} (1000 years)	mg/kg	---	7.89×10^{-5}	3.98×10^{-5}	3.07×10^{-4}

$PEC_{porewater}$: concentration in porewater.

$C_{soil}(0)$: initial concentration of the substance in soil (after the waste amendment).

Table S8. The risk characterization ratios estimated for soil organisms (RCR_{soil}), for worm-eating predators ($RCR_{oral,worm}$), for freshwater (RCR_{water}) and sediment (RCR_{sed}) organisms and for fish-eating predators ($RCR_{oral,fish}$).

	B-1	**B-2**	**B-3**	**B-4**
PFOS				
RCR_{soil}	1.49×10^{-3}	9.99×10^{-4}	1.52×10^{-3}	7.55×10^{-5}
$RCR_{oral,worm}$	0.132	0.088	0.135	0.007
RCR_{water}	1.30×10^{-3}	8.72×10^{-4}	1.07×10^{-3}	3.26×10^{-3}
RCR_{sed}	3.83×10^{-3}	2.57×10^{-3}	3.15×10^{-3}	9.62×10^{-3}
$RCR_{oral,fish}$	0.012	0.008	0.010	0.031

PFOA				
RCR_{soil}	1.47 x 10^{-5}	1.24 x 10^{-5}	8.71 x 10^{-6}	1.15 x 10^{-5}
$RCR_{oral,worm}$	1.77 x 10^{-3}	1.49 x 10^{-3}	1.04 x 10^{-3}	1.38 x 10^{-3}
RCR_{water}	1.17 x 10^{-4}	1.66 x 10^{-4}	8.41 x 10^{-5}	8.56 x 10^{-5}
RCR_{sed}	4.50 x 10^{-5}	6.38 x 10^{-5}	3.23 x 10^{-5}	3.29 x 10^{-5}
$RCR_{oral,fish}$	9.37 x 10^{-5}	1.33 x 10^{-4}	6.73 x 10^{-5}	6.85 x 10^{-5}
Penta-BDE				
RCR_{soil}	7.08 x 10^{-5}	2.05 x 10^{-4}	1.55 x 10^{-4}	4.24 x 10^{-4}
$RCR_{oral,worm}$	0.026	0.075	0.057	0.155
RCR_{water}	7.90 x 10^{-5}	5.93 x 10^{-5}	7.77 x 10^{-5}	6.41 x 10^{-5}
RCR_{sed}	2.13 x 10^{-5}	1.60 x 10^{-5}	2.10 x 10^{-5}	1.73 x 10^{-5}
$RCR_{oral,fish}$	2.65 x 10^{-3}	1.99 x 10^{-3}	2.60 x 10^{-3}	2.15 x 10^{-3}
Deca-BDE				
RCR_{soil}	4.51 x 10^{-6}	1.22 x 10^{-5}	1.23 x 10^{-5}	2.27 x 10^{-5}
$RCR_{oral,worm}$	0.006	0.016	0.016	0.029
RCR_{water}	1.38 x 10^{-3}	6.36 x 10^{-4}	1.89 x 10^{-3}	2.50 x 10^{-3}
RCR_{sed}	2.80 x 10^{-4}	1.29 x 10^{-4}	3.83 x 10^{-4}	5.09 x 10^{-4}
$RCR_{oral,fish}$	4.59 x 10^{-9}	2.12 x 10^{-9}	6.29 x 10^{-9}	8.35 x 10^{-9}
DP				
RCR_{soil}	4.63 x 10^{-4}	1.03 x 10^{-3}	5.67 x 10^{-4}	1.27 x 10^{-3}
$RCR_{oral,worm}$	1.39 x 10^{-4}	3.09 x 10^{-4}	1.71 x 10^{-4}	3.81 x 10^{-4}
RCR_{water}	---	---	---	---
RCR_{sed}	---	---	---	---
$RCR_{oral,fish}$	2.54 x 10^{-9}	2.92 x 10^{-9}	2.40 x 10^{-9}	5.26 x 10^{-9}
DBDPE				
RCR_{soil}	---	4.24 x 10^{-9}	2.14 x 10^{-9}	1.65 x 10^{-8}
$RCR_{oral,worm}$	---	0.067	0.034	0.260
RCR_{water}	6.22 x 10^{-6}	4.64 x 10^{-5}	2.70 x 10^{-4}	6.05 x 10^{-4}
RCR_{sed}	8.09 x 10^{-8}	6.04 x 10^{-7}	3.50 x 10^{-6}	7.87 x 10^{-6}
$RCR_{oral,fish}$	4.20 x 10^{-10}	3.14 x 10^{-9}	1.82 x 10^{-8}	4.09 x 10^{-8}

Table S9. Risk characterization ratios obtained for soil organisms due to the biosolid annual application to soil.

	B-1	B-2	B-3	B-4
PFOS				
RCR_{soil} (1 year)	0.002	9.99 x 10^{-4}	0.002	7.55 x 10^{-5}

RCR_{soil} (10 year)	0.004	0.003	0.004	1.95×10^{-4}
RCR_{soil} (25 year)	0.004	0.003	0.004	1.95×10^{-4}
RCR_{soil} (50 year)	0.004	0.003	0.004	1.95×10^{-4}
RCR_{soil} (100 year)	0.004	0.003	0.004	1.95×10^{-4}
PFOA				
RCR_{soil} (1 year)	1.47×10^{-5}	1.24×10^{-5}	8.71×10^{-6}	1.15×10^{-5}
RCR_{soil} (10 year)	3.49×10^{-4}	2.94×10^{-4}	2.06×10^{-4}	2.73×10^{-4}
RCR_{soil} (25 year)	3.49×10^{-4}	2.94×10^{-4}	2.06×10^{-4}	2.73×10^{-4}
RCR_{soil} (50 year)	3.49×10^{-4}	2.94×10^{-4}	2.06×10^{-4}	2.73×10^{-4}
RCR_{soil} (100 year)	3.49×10^{-4}	2.94×10^{-4}	2.06×10^{-4}	2.73×10^{-4}
Penta-BDE				
RCR_{soil} (1 year)	7.08×10^{-5}	2.05×10^{-4}	1.55×10^{-4}	4.24×10^{-4}
RCR_{soil} (10 year)	5.49×10^{-4}	0.002	0.001	0.003
RCR_{soil} (25 year)	9.09×10^{-4}	0.003	0.002	0.005
RCR_{soil} (50 year)	0.001	0.003	0.002	0.006
RCR_{soil} (100 year)	0.001	0.003	0.002	0.007
Deca-BDE				
RCR_{soil} (1 year)	4.51×10^{-6}	1.22×10^{-5}	1.23×10^{-5}	2.27×10^{-5}
RCR_{soil} (10 year)	4.10×10^{-5}	1.10×10^{-4}	1.12×10^{-4}	2.06×10^{-4}
RCR_{soil} (25 year)	8.62×10^{-5}	2.32×10^{-4}	2.36×10^{-4}	4.34×10^{-4}
RCR_{soil} (50 year)	1.33×10^{-4}	3.57×10^{-4}	3.63×10^{-4}	6.69×10^{-4}
RCR_{soil} (100 year)	1.71×10^{-4}	4.61×10^{-4}	4.68×10^{-4}	8.63×10^{-4}
RCR_{soil} (250 year)	1.87×10^{-4}	5.03×10^{-4}	5.10×10^{-4}	9.40×10^{-4}
RCR_{soil} (500 year)	1.87×10^{-4}	5.04×10^{-4}	5.11×10^{-4}	9.42×10^{-4}
DP				
RCR_{soil} (1 year)	4.63×10^{-4}	0.001	5.67×10^{-4}	0.001
RCR_{soil} (10 year)	0.005	0.010	0.006	0.012
RCR_{soil} (25 year)	0.011	0.023	0.013	0.029
RCR_{soil} (50 year)	0.019	0.042	0.023	0.052
RCR_{soil} (100 year)	0.031	0.070	0.038	0.086
RCR_{soil} (250 year)	0.048	0.107	0.059	0.132
RCR_{soil} (500 year)	0.054	0.120	0.066	0.145
RCR_{soil} (750 year)	0.055	0.0121	0.067	0.150
DBDPE				

RCR_{soil} (1 year)	---	4.24×10^{-9}	2.14×10^{-9}	1.65×10^{-8}
RCR_{soil} (10 year)	---	4.10×10^{-8}	2.07×10^{-8}	1.60×10^{-7}
RCR_{soil} (25 year)	---	9.64×10^{-8}	4.87×10^{-8}	3.75×10^{-7}
RCR_{soil} (50 year)	---	1.74×10^{-7}	8.80×10^{-8}	6.79×10^{-7}
RCR_{soil} (100 year)	---	2.89×10^{-7}	1.46×10^{-7}	1.12×10^{-6}
RCR_{soil} (250 year)	---	4.44×10^{-7}	2.24×10^{-7}	1.71×10^{-6}
RCR_{soil} (500 year)	---	4.98×10^{-7}	2.51×10^{-7}	1.94×10^{-6}
RCR_{soil} (750 year)	---	5.04×10^{-7}	2.55×10^{-7}	1.96×10^{-6}
RCR_{soil} (1000 year)	---	5.05×10^{-7}	2.55×10^{-7}	1.97×10^{-6}

Table S10. Comparison of concentrations of PFOS, PFOA, Penta-BDE, Deca-BDE, DP and DBDPE in different environmental compartments.

Compound	Compartment	Concentration	Reference
PFOS			
	Biosolid (ng/g d.w.)	3.3 - 54.1	Guo et al., 2010
		1.56 - 36.6	Li et al., 2010
		35.5 - 46.2	Esparza et al., 2011
		N.D. - 26	Roig et al., 2012
		4.2 - 169	Sun et al., 2012
		20 - 175	Yan et al., 2012
		16-110	Perkola and Sainio, 2013
		69	Zareitalabad et al., 2013
		41.4[a]	Campo et al., 2014
		12 - 50[a]	Ulrich et al., 2016
		0.65-64.4	Values used in the ERA
	WWTP Effluent (ng/L)	<0.06 - 82.2	Ahrens et al., 2009
		2.56 - 3.85	Sun et al., 2011
		<0.18 - 21.0	Arvaniti et al., 2012
		<10 - 200	Kim et al., 2012
		10-1000	Zareitalabad et al., 2013
		3.8-1300	Perkola and Sainio, 2013
		219-819[d]	Values used in the ERA

Crop grown in biosolid amended soils (ng/g)	< 0.05 - 0.7 [a, c] (potato)	Lechner and Knapp, 2011
	0.5-18.4 [a, c] (carrot)	Lechner and Knapp, 2011
	N.D.-1.3 [a, c] (cucumber)	Lechner and Knapp, 2011
	101.62[a,b] (lettuce)	Blaine et al., 2013
	<0.14[a,b] (tomato)	Blaine et al., 2013
	0.80-2.53[a, c] (wheat)	Wen et al., 2014
	N.D. - 0.003[c] (tomato)	Values used in the HRA
PFOA		
Biosolid (ng/g d.w.)	7.0 - 30.3	Llorca et al., 2011
	8.0 - 68	Sepulvado et al., 2011
	N.D. - 20	Sun et al., 2011
	N.D. - 23	Roig et al., 2012
	0.65-0.91	Perkola and Sainio, 2013
	1.83[a]	Campo et al., 2014
	0.32 - 16[a]	Ulrich et al., 2016
	2.52-9.35	Values used in the ERA
WWTP Effluent (ng/L)	12.3 -77.6	Ahrens et al., 2009
	18.4 -41.1	Sun et al., 2011
	<0.72 - 34.0	Arvaniti et al., 2012
	<10 - 3000	Kim et al., 2012
	10-1000	Zareitalabad et al., 2013
	6.6-100	Perkola and Sainio, 2013
	16.8-33.2[d]	Values used in the ERA
Crop grown in biosolid amended soils (ng/g)	2.9-7.7 [a, c] (potato)	Lechner and Knapp, 2011

	30.8-31.3 [a, c] (carrot)	Lechner and Knapp, 2011
	11.3-23.8 [a, c] (cucumber)	Lechner and Knapp, 2011
	20.01[a, b] (lettuce)	Blaine et al., 2013
	<0.14[a, b] (tomato)	Blaine et al., 2013
	1.01-2.90[a, c] (wheat)	Wen et al., 2014
	N.D. - 0.014[c] (tomato)	Values used in the HRA
Penta-BDE		
Biosolid (ng/g d.w.)	61.4-245	Andrade et al., 2010
	N.D. - 5.9	Yang et al., 2011
	1.3 - 48.7	Cincinelli et al., 2012
	135-810	Davis et al., 2012
	4.4 - 16	Olofsson et al., 2012
	N.D. - 180	Roig et al., 2012
	N.D. - 10.3	Barón et al., 2014
	180-730[a]	Gaylor et al., 2014
	1.82-17.3	Values used in the ERA
WWTP Effluent (ng/L)	0.35-68	Daso et al., 2012
	9.04	Hope et al., 2012
	N.D. -2.21	Jiang et al., 2018
	0.01-0.02	Xiang et al., 2013
	N.D.	Schreder and La Guardia, 2014
	0.1-7.0	Deng et al., 2015
	54	Gallen et al., 2016
	0.34-0.46[d]	Values used in the ERA
Crop grown in biosolid amended/contaminated soils (ng/g)	4 (zucchini)	Mueller et al ., 2006

	0.7-2.1[b] (nightshade)	Vrkoslavová et al., 2010
	0.6-1.2[b] (lettuce)	Wang et al., 2011
	N.D. -0.3[b] (radish)	Wang et al., 2011
	N.D. (corn)	Hale et al., 2012
	0.007-0.01[c] (tomato)	Values used in the HRA
Deca-BDE		
Biosolid (ng/g d.w.)	792-1220	Andrade et al., 2010
	50 - 190	Clarke et al., 2010
	5.45-843	de la Torre et al., 2012
	636-2933	Davis et al., 2012
	510	Olofsson et al., 2012
	10 - 1400	Roig et al., 2012
	539[a]	Gorga et al., 2013
	< 0.67 - 319	Barón et al., 2014
	950[a]	Gaylor et al., 2014
	29.3-347	Values used in the ERA
WWTP Effluent (ng/L)	< 2.4 - 9770	Daso et al., 2012
	17.2	Hope et al., 2012
	1.06-1.72	Xiang et al., 2013
	2.4-22.2	Schreder and La Guardia, 2014
	N.D. - 31	Deng et al., 2015
	9.5-68.6	Li et al., 2018
	92.4-364[d]	Values used in the ERA
Crop grown in biosolid amended/contaminated soils (ng/g)	N.D. (nightshade)	Vrkoslavová et al., 2010
	4.9-8.4[b] (lettuce)	Wang et al., 2011

	0.6[b] (radish)	Wang et al., 2011
	N.D. (corn)	Hale et al., 2012
	0.671[c] (tomato)	Values used in the HRA
DP		
Biosolid (ng/g d.w.)	0.103 - 0.263	Qi et al., 2010
	2.45-93.8	de la Torre et al., 2011
	3.0-39	Davis et al., 2012
	2.66-20	Barón et al., 2014
	2.28-14.6	Values used in the ERA
WWTP Effluent /Landfill leachate (ng/L)	0.03-2.39	Qi et al., 2010
	0.17-26.3	Zheng et al., 2011
	<0.01 - 1.2	Li et al., 2018
	13.6-29.8[d]	Values used in the ERA
Plant near contaminated area (ng/g)	0.45-16.7 (eucalyptus foliage)	Chen et al., 2011
	0.51-51.9b (pine needle)	Chen et al., 2011
	0.007-0.011[c] (tomato)	Values used in the HRA
DBDPE		
Biosolid (ng/g d.w.)	3.25-125	de la Torre et al., 2012
	81.0[a]	Gorga et al., 2013
	1.9-6.3	Nyholm et al., 2013
	<12.09 - 124	Barón et al., 2014
	0.10-0.15	Values used in the ERA
WWTP Effluent (ng/L)	<0.06 - 7.14	Kim et al., 2014

5.61[a]	Li et al., 2016
<0.35 - 21.8	Li et al., 2018
0.06-5.58[d]	Values used in the ERA

N.D.: not detected. ERA: Environmental risk assessment. HRA: Healthy risk assessment.

[a]Mean values. [b]Concentrations expressed in ng/g d.w. [c]Concentrations expressed in ng/g w.w. [d]Concentrations in runoff water from biosolid amended soils

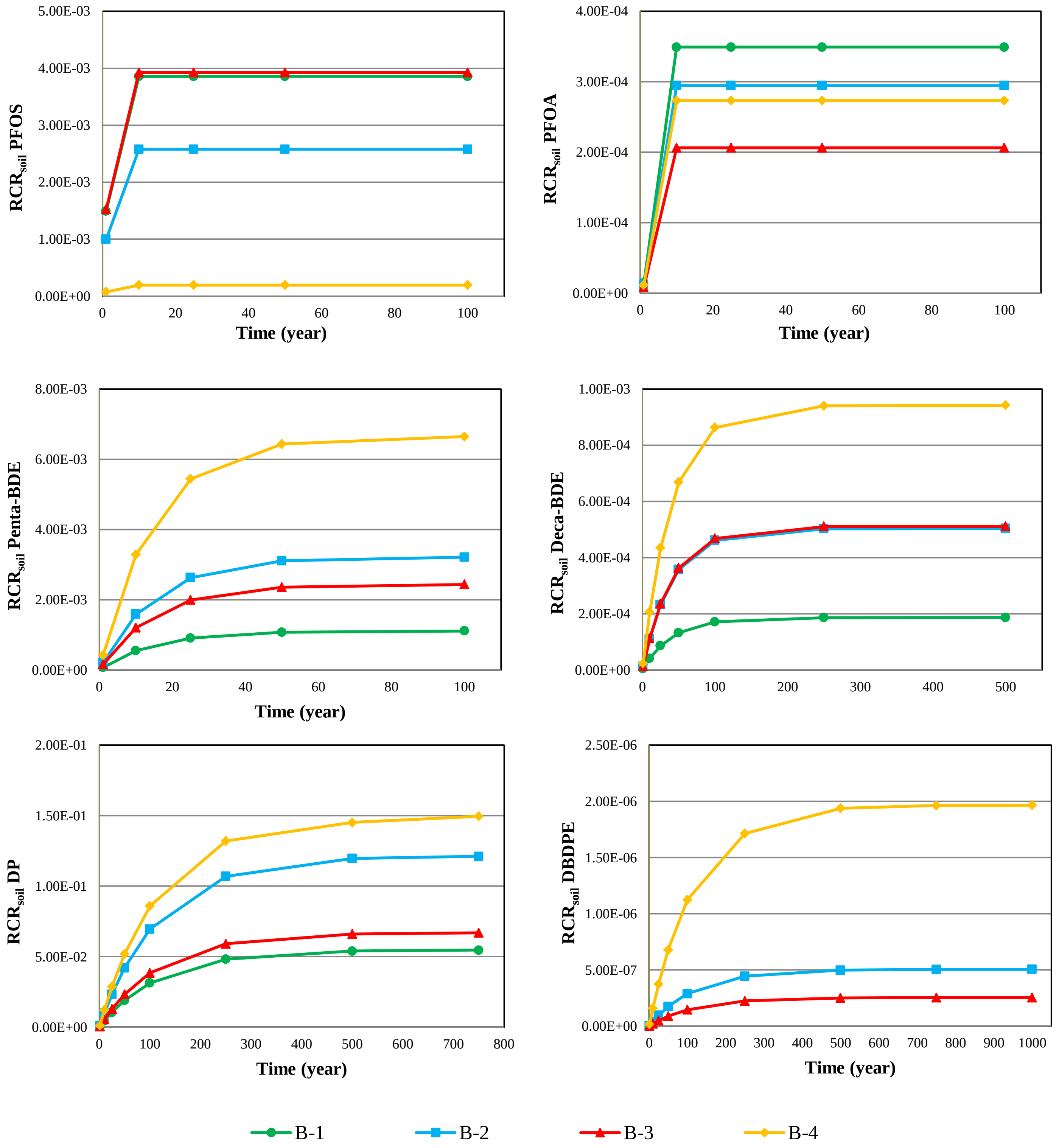

Fig. S1.- Evolution of the risk characterization ratios for soil organisms due to the biosolid annual application to soil.

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