Supplementary Information to

PERSISTENT ORGANIC POLLUTANTS IN BOREAL AND MONTANE SOIL PROFILES: DISTRIBUTION, EVIDENCE OF PROCESSES AND IMPLICATIONS FOR GLOBAL CYCLING

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Text SI-1: Differentiation of different layers of the organic horizon.

The organic (O-) horizon can be divided into three layers according to the degree of decomposition of the organic matter:

Oi: refers to the litter layer, consisting of mostly undegraded plant material (< 10 % decomposed material),

Oe: refers to the fermentation layer, consisting of fragments of plant material (origin easily recognizable) but also 10 to 70% decomposed organic material,

Oa: refers to the humous layer, consisting mostly (> 70%) of decomposed organic matter whose origin is not recognizable anymore.

Text SI-2: Description of extraction and clean-up methods for soil samples

Norwegian samples: Samples were mixed with anhydrous sodium sulfate and Soxhlet extracted for 16 h using dichloromethane, after being spiked with a recovery standard containing $^{13}C_{12}$ labeled PCB congeners 28, 52, 101, 138, 153, 180 and 209 and PBDE congeners 51, 128 and 190. The extracts were cleaned on activated alumina/silica columns, followed by treatment with concentrated sulfuric acid and gel permeation chromatography (GPC). After solvent exchange to dodecane (final volume 25 µL, containing internal standards PCB-30, $^{13}C_{12}$ PCBs -141 and -208, BDEs -69, and -181) they were analyzed for a total of 44 target PCBs (congener numbers 18, 22, 28+31, 41+64, 44, 49, 52, 54, 70, 74, 87, 90+101, 95, 99, 104, 105, 110, 114, 118, 123, 132+153, 138, 141, 149, 151, 155, 156, 157, 158, 167, 170, 174, 180, 183, 187, 188, 189, 194, 199, 203), 18 target PBDEs (congener numbers 17, 28, 35, 37, 47, 49, 66, 71, 77, 85, 99, 100, 119, 138, 153, 154, 166, 183), p,p'-DDD, -DDE, DDT, o,p'-DDD, -DDE, -DDT and the recovery compounds on a Finnigan Trace GC-MS operated in positive electron ionization mode using selected ion monitoring. A CP-SIL 8CB column was used (50 m, 0.25 mm i.d., 0.12 µm film thickness, Varian) with helium as the mobile phase at a constant flow of 1 mL/min. Quantification of α-, ß- and γ-HCH was performed on the same instrument operated in negative chemical ionization mode with ammonia as the reagent gas using selected ion monitoring.

Italian samples: Freeze-dried, homogenized samples were Soxtech-extracted (SER 148 all glass-Soxtech, Velp Scientifica, Usmate, Italy) for 6 h using n-hexane/acetone 6:1 after being spiked with a PCB recovery standard containing congeners 40 and 128. Clean-up of the extracts included GPC and fractionation on activated Florisil columns. Cleaned extracts were reduced to 50 µL after addition of internal standards (PCB-30) and analyzed for a total of 32 target PCBs (28+31, 52, 44, 49, 64, 74, 70, 104, 101, 99, 97, 87, 110, 118, 105, 136, 151, 149, 132+153, 138, 156, 188, 187, 183, 174, 177, 180, 201, 203, 194), p,p'-DDD -DDT, -DDE, o,p'-DDD -DDT, -DDE. Analyses were performed using a Hewlett-Packard 5890 series 2 gas chromatograph equipped with dual ECD. Injection was split via a Y connector into two parallel columns: a 60m DB-5 (J&W Scientific, Folsom, CA, i.d. 0.25 mm, film thickness 0.25 µm) and a 60 m BP-50 (SGE International, Melbourne, Australia, i.d. 0.25 mm, film thickness 0.25 μm). Carrier gas (He) flow rate was 1 mL/min.

Text SI-3: Estimation of PCB fractions lost by leaching and degradation per year.

Fractions of PCBs leached as truly dissolved compounds or in association with dissolved organic matter (DOM) were estimated based on annual amounts of precipitation. Since no site specific precipitation data was available, typical ranges for the areas were used, resulting in a minimum and maximum estimate at each site for individual congeners. The Italian sites are located in an area that receives 750 – 1250 mm of precipitation per year on average and all Norwegian sites receive 1250 – 2500 mm per year (1). To obtain the amount of water which typically percolates through the top layers of the soil profile every year (F_W, g m^{-2} $year^{-1}$), these precipitation amounts were reduced by estimated interception evaporation (23% at site I-1400 and 12.5% at site I-1800 and all Norwegian sites, according to data obtained in similar forest types with similar amounts of rainfall (2)), and soil evaporation (ca. 60 mm year-1 (3)). This approach used to account for precipitation that does not percolate through the soil profile is probably not very accurate as it may vary between sites. However, given the wide range of annual precipitation used for this estimation that uncertainty should not affect the results too much.

In all profiles except for I-1100 the organic horizon contained the main portion of heavy PCBs which are believed not to experience extensive translocation (see discussion in the main text). Therefore fractions of PCBs leached downward were calculated for this horizon only. I-1100 was excluded from this exercise since the homogenous POP distribution within the profile suggested that bioturbation plays a much more important role in translocating POPs here.

To estimate the fraction of PCBs leached out of the O horizons as dissolved phase compounds ($f_{leach\text{-}diss}$) every year, they were assumed to reach equilibrium between SOM (expressed as organic carbon) and percolating water:

$$f_{leach-diss} = \frac{F_W}{C_{org-O} \cdot K_{OC}} \qquad 1)$$

where $C_{org\text{-}O}$ is the amount of organic carbon stored in the O horizon (g m^{-2}) and K_{OC} is the organic carbon-water partition coefficient that was estimated according to (4):

$$K_{OC} = 10^{0.456 + 0.72 \cdot \log K_{OW}} \qquad 2)$$

with temperature adjusted K_{OW} taken from Li et al. (5).

To estimate the fraction of PCBs leached out of the O horizon in association with DOM (expressed as dissolved organic carbon (DOC)) ($f_{leach\text{-}DOC}$), most DOM was assumed to originate from SOM (6). Thus leaching of DOC was regarded as a loss of soil organic carbon including the POPs bound to it. Fluxes of DOC (F_{DOC}) leached out of the organic horizon were estimated from F_W and a DOC concentration (c_{DOC}) of 40 mg L^{-1} as a typical DOC concentrations observed in leachates from the O horizon in several different studies conducted in regions with similar environmental conditions (6).

$$f_{leach-DOC} = \frac{F_W \cdot c_{DOC} \cdot K_{DOC}}{C_{org-O} \cdot K_{OC}} \qquad 3)$$

where K_{DOC} is the DOC-water partition coefficient that was estimated according to (7):

$$K_{DOC} = 10^{-0.5 + 0.71 \cdot \log K_{OW}} \qquad 4).$$

The small fraction of SOM that is prone to turn into DOM was believed to be more polar than "average" SOM and thus the weight specific amounts of POPs bound to DOM should be lower than those bound to "average" SOM. Multiplication with the ratio between K_{DOC} and K_{OC} accounts for this difference.

The results of this estimation are shown in Table SI-5a. $f_{leach\text{-}diss}$ and $f_{leach\text{-}DOC}$ (expressed as %) should be regarded as fractions based on the amount of PCBs present in the organic layers at a given time. Minimum and maximum values result from minimum and maximum yearly precipitation amounts, respectively. The approach used assumes homogenous distribution of POPs within the O horizon as a simplification. However, the wide range of yearly precipitation amounts is believed to account for the highest uncertainty but the range between minimum and maximum f_{leach} still gives a useful idea of the extent of leaching of PCBs in the soil profiles investigated. Table SI-5b shows estimates of temperature corrected PCB fractions lost from soil by degradation (f_{degrad}, % year^{-1}) (8). These values were recommended for modelling the fate and behaviour of PCBs in the environment (8) but have not been validated for environmental conditions.

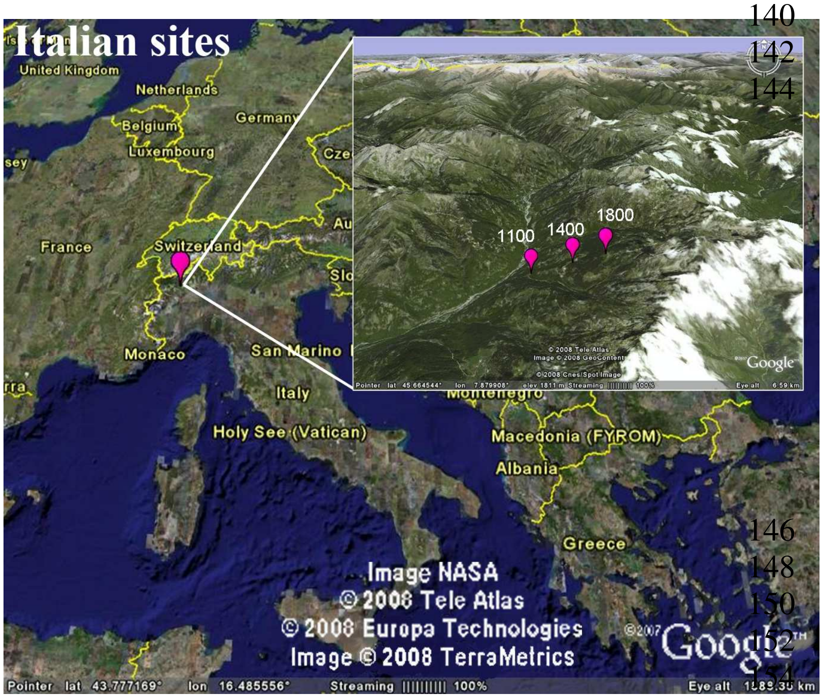
Text SI-4: Estimation of the Oi layer age and amounts of PCB-180 deposited to these layers.

The average residence time of organic matter in the litter layer (Oi) at the Italian sites could be estimated from the area specific mass of litter [g m^{-2}] (calculated as the product of thickness [cm] and bulk density [g cm^{-3}] of these layers (Table SI-1)), divided by yearly litter deposition rates [g m^{-2}]. Calculation of litter deposition based on forest canopy biomass measurements (method described by Nizzetto et al. (9)), assumed constant biomass production during the last decade. The biomass at the end of the growing season was derived from the ratio of the leaf area index (LAI) and the specific leaf area (SLA). LAI were 3.9, 4.8 and 1.7 m^2 m^{-2}, SLA were 0.014, 0.030 and 0.011 m^2 g^{-1} dw at sites I-1100, I-1400 and I-1800, respectively. The average residence time of organic matter in the Oi layer at sites I-1100, I-1400 and I-1800 was estimated at 2.8, 8.5 and 7.7 years, respectively.

The load U_d [μg m^{-2}] of PCB-180 deposited to the Oi layer was estimated considering the annual deposition reported by Nizzetto et al. (10) for the same sites (assumed to be constant during the last decade) multiplied by the average residence time of the O_i horizon. Using the temperature corrected degradation rate reported for PCB-180 (8), the loss due to reaction in a period of 10 years can be considered negligible. U_d of deeper horizons could not be estimated due to the lack of information on deposition rates in the past.

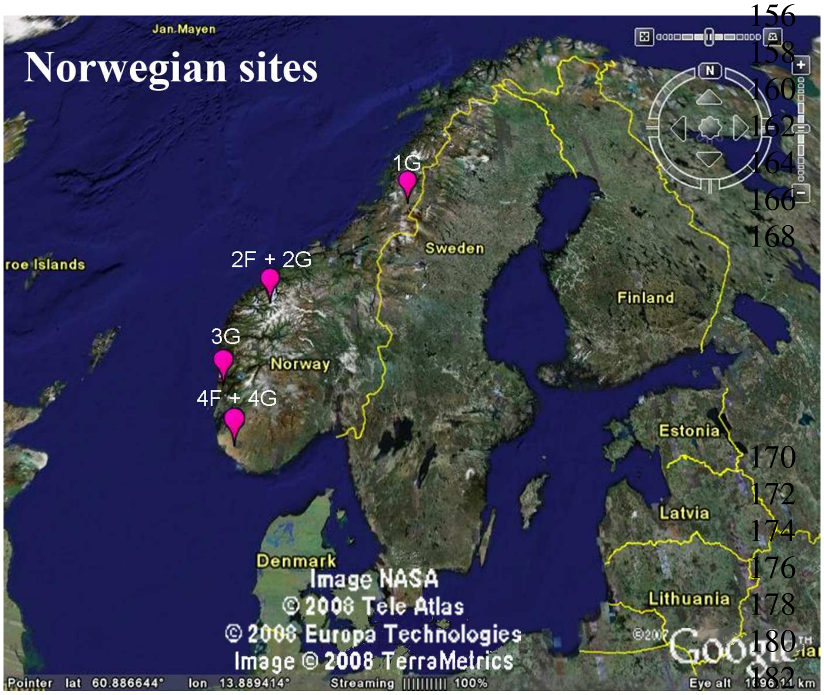
It should be noted that both the absolute amount U of chemicals in the Oi layer as well as U_d depend on the bulk density (BD) estimated for the Oi layers. However, varying BD does not affect the conclusions drawn, as it results in changing both U and U_d by the same factor.

Figure SI-1: Location of sample sites.
(Maps obtained from Google Earth 4.3[11].)



Overview map[1]
centre latitude: 44 °
centre longitude: 16 °
eye altitude[2]: 1888 km

Detailed map[1]
center latitude: 45.662 °
center longitude: 7.882 °
eye altitude[2]: 6.6 km



Map[1]
centre latitude: 60 °
centre longitude: 20 °
eye altitude[2]: 1696 km

[1] ref. 11
[2] "Eye altitude" is used in Google Earth 4.3 to indicate how far was zoomed in to obtain the pictures shown.

Figure SI-2: Fractions of individual PCB congeners as a proportion of the total measured in the different soil profiles, plotted against their log K_{OW}. a) Italian cores; b) Norwegian cores.

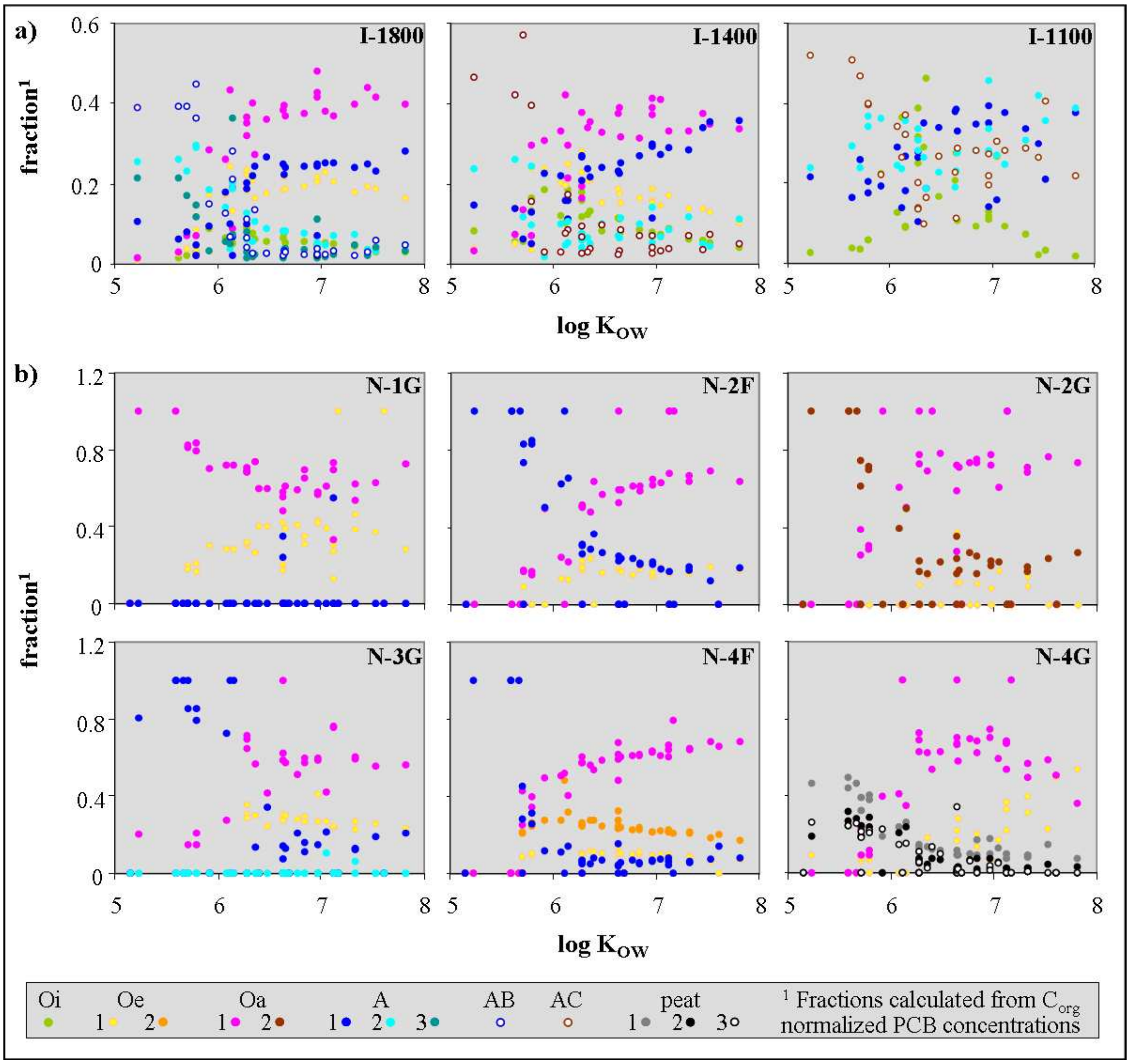


Figure SI-3: Organic matter normalized concentrations of selected PCBs in organic compartments (including vegetation and superficial litter for the Italian profile) against C/N-ratios.

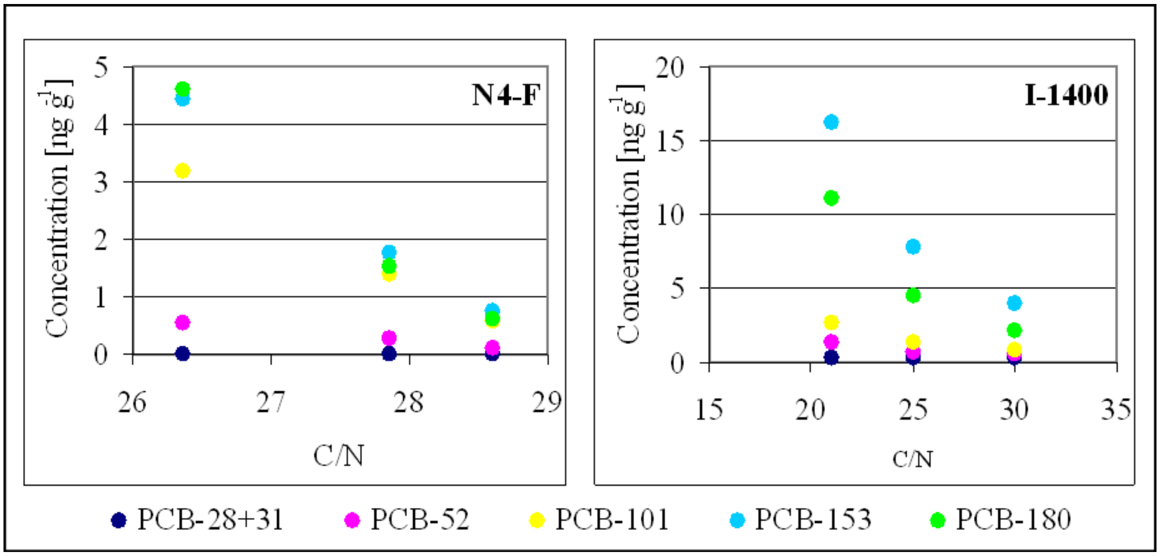


Table SI-1: Location of the soil profiles and soil property details.

profile	latitude [°]	longitude [°]	horizon	layer thickness [cm]	C_{org} [%][a]	C/N	bulk density[b] [g cm^{-3}]	pH
			Oi	2.3	43	22	0.05[c]	n.d.
			Oe	1.2	26	20	0.27	n.d.
			Oa	0.5	16	18	0.40	n.d.
I-1800	45.6390	7.88823	A1	2.0	7.3	17	0.65	4
			A2	8.0	3.7	21	0.90	4.3
			A3	10	2.9	25	0.99	4.4
			AB	10	2.1	29	1.10	4.7
			Oi	2.7	44	30	0.05[c]	n.d.
			Oe	3.3	39	25	0.19	n.d.
I-1400	45.6395	7.87803	Oa	1.0	19	21	0.34	n.d.
			A1	5.0	10	18	0.52	3.7
			A2	5.0	5	25	0.75	4.2
			AC	10.0	3.4	25	0.93	4.5
			Oi	1.5	40	30	0.05[c]	n.d.
I-1100	45.6408	7.86463	A1	10	5.8	14	0.73	5.5
			A2	10	3.4	14	0.93	5.3
			AC	10	3.0	15	0.98	5.7
			Oe	1.0	52	32	0.15	4.4
N-1G	64.97	13.58	Oa	1.0	48	28	0.16	4.2
			A	2.0	22	24	0.31	4.6
			Oe	1.0	50	38	0.15	4.7
N-2F	62.02	6.69	Oa	2.0	46	25	0.16	4.4
			A	1.5	13	22	0.47	4.5
			Oe	2.0	49	34	0.15	4.4
N-2G	62.02	6.69	Oa1	2.0	51	24	0.15	4.1
			Oa2	2.0	52	21	0.15	4.2
			Oe	2.0	51	35	0.15	4.7
N-3G	59.88	5.32	Oa	2.0	48	25	0.16	4.8
			A	2.0	15	19	0.41	4.9
			A	2.0	21	19	0.33	4.9
			Oe1	1.0	49	29	0.15	4.3
N-4F	58.55	6.37	Oe2	1.0	49	28	0.15	4.0
			Oa	1.0	46	26	0.16	3.8
			A	1.0	40	26	0.19	4.0
			Oe	2.0	49	20	0.15	4.3
			Oa	2.0	47	20	0.16	4.4
N-4G	58.55	6.37	peat1	2.0	48	19	0.16	4.4
			peat2	2.0	53	26	0.14	4.1
			peat3	2.0	54	25	0.14	4.0

[a] % of dry weight

[b] estimated according to ref 12

[c] bulk density of Oi layer was estimated at 0.05 g cm^{-3}, as Federer (13) suggests overestimation when using the equation given by Huntington et al. (12)

Table SI-2: Concentration [pg g^{-1}] of selected PCBs in superficial litter samples.

Concentrations are given on a dry weight basis.

1800 m[1]

sampling date	PCB 28/31	PCB 52	PCB 101	PCB 138	PCB 153	PCB 180
09.05.2005	340	730	680	2300	2200	1200
20.06.2005	240	670	690	2300	2300	1400
05.09.2005	280	370	470	1800	2100	1300
17.10.2005	460	710	720	2500	2400	1500

1400 m

sampling date	PCB 28/31	PCB 52	PCB 101	PCB 138	PCB 153	PCB 180
25.04.2005	330	560	780	2300	2200	1100
09.05.2005	190	350	490	1400	1400	690
20.06.2005	270	630	620	2700	2600	1500
05.09.2005	310	430	640	2500	2500	1400
17.10.2005	360	580	790	2400	2400	1400

1100 m

sampling date	PCB 28/31	PCB 52	PCB 101	PCB 138	PCB 153	PCB 180
25.04.2005	580	750	1100	1800	1800	780
09.05.2005	450	690	930	1800	1700	780
20.06.2005	650	980	900	1900	2400	1100
05.09.2005	330	660	890	1700	2200	980
17.10.2005	660	860	1100	2700	2900	1500

[1] During the first sampling campaign no superficial litter sample could be taken at site I-1800 as the ground was still covered with snow.

Table SI-3a: Chemical concentration data (pg g^{-1} dw) Italian profiles – PCBs and OCPs.

	Profile / Layers I-1800						
	Oi	Oe	Oa	A1	A2	A3	AB
PCB/OC							
18	45	<28	<28	51	63	42	54
28+31	90	96	62	62	130	84	110
49	65	130	66	<17	64	25	45
52	160	170	190	100	150	86	140
64	45	31	<22	<22	36	<22	30
66	280	410	360	58	90	25	70
70	810	1300	1200	380	150	47	78
74	190	180	120	13	61	92	39
87	440	670	660	160	28	<20	<20
95	350	430	420	65	65	<16	29
97	180	270	220	31	<20	<20	<20
99	360	590	590	240	54	<19	39
101	360	640	740	200	64	<12	18
105+132	690	1200	1300	350	31	<29	<29
110+151	680	1100	1300	330	42	<22	<22
118	810	1400	1800	520	88	<27	<27
136	160	360	380	40	<16	<16	<16
138	1900	4200	4900	1500	240	85	40
149	910	1800	2200	760	130	29	23
153	2100	4600	5400	1600	280	100	57
156	470	1100	1200	390	56	<28	<28
174	490	1400	1800	460	41	<22	<22
177	280	770	940	260	<22	<22	<22
180	1200	3100	4200	1200	180	88	30
183	480	1300	1300	400	38	<21	<21
187	430	1300	1700	110	43	72	<22
194	190	640	930	300	25	<14	<14
201	250	900	1200	310	23	<22	<22
203	120	480	640	170	<25	<25	<25
α-HCH	200	150	140	<23	<23	<23	<23
γ-HCH	300	270	130	54	40	30	40
o,p'-DDD	140	270	380	99	<9	<9	<9
p,p'-DDD	850	1300	1900	390	53	<11	<11
o,p'-DDE	62	81	100	37	<14	<14	<14
p,p'-DDE	2300	6251	9400	3300	610	160	65
o,p'-DDT	750	1600	2100	660	120	42	<28
p,p'-DDT	2600	6000	7800	1700	340	110	57
p,p'-DDT/p,p'-DDE	0.83	0.52	0.55	0.68	0.88	n.a.	n.a.

Table SI-3a continued.

	Profile / Layers									
	I 1400						I 1100			
	Oi	Oe	Oa	A1	A2	AC	Oi	A1	A2	AC
PCB/OC										
18	79	32	<28	34	29	35	91	110	71	140
28+31	100	86	62	62	62	62	180	62	62	180
49	120	160	96	<17	<17	<17	110	59	57	59
52	240	240	260	64	64	190	250	280	150	260
64	47	<22	<22	<22	<22	<22	65	29	35	33
66	300	410	240	70	<25	<25	200	85	59	53
70	700	1000	840	300	73	<25	500	170	84	100
74	180	180	92	26	<25	<25	130	38	43	40
87	340	470	140	120	<20	<20	310	37	23	<20
95	340	390	250	98	<8	<8	360	45	49	26
97	220	280	120	89	<20	<20	260	<20	17	<20
99	400	560	470	160	19	<19	1300	76	45	35
101	350	520	490	150	29	<12	380	61	43	15
105+132	630	870	960	300	46	<29	460	94	33	<29
110+151	580	860	700	270	38	<22	440	77	34	<22
118	800	1150	1100	390	62	<27	430	110	33	35
136	140	230	250	50	<16	<16	150	<16	<16	<16
138	1700	2900	2800	1300	250	110	750	420	200	170
149	820	1350	1437	570	120	74	500	150	58	60
153	1700	3000	3100	1300	320	150	1000	450	220	200
156	420	720	700	330	51	<28	180	110	41	41
174	380	710	830	360	46	<22	180	93	39	26
177	230	450	570	230	22	<22	69	<22	21	<22
180	900	1700	2100	980	210	80	430	290	160	130
183	330	580	810	290	38	<21	130	<21	<21	<21
187	410	850	930	140	90	28	170	76	42	22
194	150	320	530	300	51	<28	<28	47	28	<28
201	230	470	640	310	25	<22	<22	24	20	<22
203	110	240	320	180	<25	<25	<25	<25	<25	<25
α-HCH	300	280	140	60	<23	<23	130	<23	<23	<23
γ-HCH	360	470	160	110	42	76	380	71	44	73
o,p'-DDD	93	140	150	71	<9	<9	22	<9	<9	<9
p,p' DDD	470	800	990	460	110	24	170	50	14	<11
o,p'-DDE	36	29	<14	28	<14	<14	42	<14	<14	<14
p,p'-DDE	1500	2500	2600	1700	500	200	790	250	160	130
o,p'-DDT	600	940	1079	440	140	71	460	80	36	40
p,p' DDT	2300	4200	5300	2000	600	300	1200	260	140	110
p,p'-DDT/p,p'-DDE	2.05	1.19	1.20	1.51	n.a.	n.a.	0.88	0.82	n.a.	n.a.

Table SI-3b: Norwegian profiles – PCBs and OCPs.

	Profile / Layers					
	N-1G			N-2F		
	Oe	Oa	A	Oe	Oa	A
PCB/OC						
18	<31	140	<40	<168	<134	220
22	<51	<122	<65	<272	<217	250
28	<130	<313	<167	<694	<554	640
31	<91	230	<118	<488	<390	400
41+64	25	88	<20	<84	150	200
44	23	89	<13	<54	120	160
49	17	82	<10	<42	84	130
52	40	170	<13	80	140	160
54	<4	<11	<6	<24	<19	<6
60+56	<56	<134	<72	<298	<238	160
70	68	160	<27	150	260	180
74	34	80	<17	91	140	120
87	41	91	<6	69	180	30
95	79	170	<26	<107	210	57
90+101	140	280	<30	200	500	81
99	52	140	<6	87	160	26
104	<4	<11	<6	<24	<19	<6
105	35	93	31	93	200	25
110	130	260	<13	210	450	62
114	<4	<11	<6	<24	<19	<6
118	94	240	48	160	480	48
123	14	18	<6	<24	39	<6
132+153	250	360	<13	250	790	86
138	400	690	<16	490	1500	140
141	78	100	<9	68	260	27
149	270	370	<27	270	870	110
151	86	120	<15	<60	250	39
155	<4	<11	<6	<24	<19	<6
156	18	37	<6	28	110	7.4
157	4.8	12	9.0	<24	30	<6
158	34	72	<6	43	110	11
167	9.9	25	<6	<24	40	<6
170	110	160	<6	130	430	35
174	120	140	<11	94	380	33
180	220	240	<12	190	740	52
183	68	97	<6	77	230	18
187	150	190	<6	140	470	44
188	<4	<11	<6	<24	<19	<6
189	5.0	<11	<6	<24	<19	<6
194	26	63	<6	39	130	10
199	6.4	<11	<6	<24	22	<6
203	38	59	<6	47	160	7.4
α-HCH	1400	2400	1800	980	540	480
β-HCH	290	840	500	240	500	140
γ-HCH	990	990	<70	1600	<220	830
o,p'-DDD	24	130	8.6	<24	65	<6
p,p'-DDD	43	290	14	37	72	13
o,p'-DDE	<61	22	<79	<330	20	<86
p,p'-DDE	<175	1200	17	320	330	110
o,p'-DDT	220	1100	<6	330	690	140
p,p'-DDT	570	2600	24	630	640	150
p,p'-DDT/p,p'-DDE	n.a.	2.26	1.39	1.96	1.90	1.43
α/β-HCH	4.81	2.89	3.61	4.01	1.08	3.58

Table SI-3b continued.

	Profile / Layers						
	N-2G			N-3G			
	Oe	Oa1	Oa2	Oe	Oa	A1	A2
PCB/OC							
18	<117	<109	440	<156	160	210	<103
22	<190	<177	310	<252	<252	270	<167
28	<485	<452	850	<644	<643	790	<425
31	<342	<318	750	<453	<453	400	<300
41+64	<59	63	150	<78	92	170	<51
44	<38	46	140	<50	<50	130	<33
49	<29	48	120	<39	98	120	<26
52	<38	100	170	<51	72	130	<33
54	<17	<16	<21	<22	<22	<10	<15
60+56	<208	<194	<260	<277	<276	200	<183
70	<79	180	120	<105	180	150	<69
74	<49	67	66	<65	<65	93	<43
87	<17	100	29	44	75	<10	<15
95	<75	120	<93	<99	<99	<46	<65
90+101	<88	320	<110	160	350	<55	<77
99	26	130	29	130	230	17	<15
104	<17	<16	<21	<22	<22	<10	<15
105	28	96	39	120	250	19	<15
110	44	330	79	84	200	<23	<32
114	<17	<16	<21	<22	<22	<10	<15
118	47	300	68	300	580	21	<17
123	31	24	30	<22	32	<10	<15
132+153	65	400	100	410	740	53	<32
138	120	840	190	830	1600	94	<41
141	<27	110	39	83	140	18	<24
149	<78	390	110	140	230	59	<69
151	<42	100	<53	<56	<56	<26	<37
155	<17	<16	<21	<22	<22	<10	<15
156	<17	40	<21	56	170	<10	<15
157	<17	<16	<21	<22	<22	<10	<15
158	<17	75	25	77	150	13	<15
167	<17	30	<21	36	100	<10	<15
170	45	220	56	210	530	35	23
174	<31	180	52	76	110	<20	<28
180	38	300	83	300	640	43	<32
183	30	110	40	110	170	27	18
187	28	260	72	210	430	33	<15
188	<17	<16	<21	<22	<22	<10	<15
189	<17	<16	<21	<22	<22	<10	<15
194	<17	79	30	73	170	19	<15
199	<17	<16	<21	<22	<22	<10	<15
203	<17	87	28	70	140	15	<15
α-HCH	430	<25	<34	<36	<36	<17	<24
β-HCH	110	180	90	260	360	490	83
γ-HCH	890	<180	<240	<260	910	<120	<170
o,p'-DDD	<17	56	<21	<22	<22	<10	<15
p,p'-DDD	33	270	90	100	190	40	<15
o,p'-DDE	<230	<210	<290	<300	<300	<140	<200
p,p'-DDE	110	980	240	710	790	110	<580
o,p'-DDT	<17	370	<21	<22	<22	<10	<15
p,p'-DDT	220	2100	360	780	1600	58	<77
p,p'-DDT/p,p'-DDE	2.00	2.12	1.52	1.10	2.08	0.53	n.a.
α/ß-HCH	4.03	0.47	n.a.	0.70	0.82	0.19	0.44

Table SI-3b continued.

	Profile / Layers								
	N-4F				N-4G				
	Oe1	Oe2	Oa	A	Oe	Oa	peat1	peat2	peat3
PCB/OC									
18	<80	<77	<72	160	120	<60	550	220	340
22	<129	<124	<117	190	<132	<97	360	260	220
28	<329	<317	<299	500	<337	<247	860	520	530
31	<232	<223	<211	300	<237	<174	740	400	410
41+64	51	150	180	140	<41	46	200	140	120
44	30	73	83	130	33	45	180	110	110
49	54	120	190	110	33	54	170	110	100
52	52	130	250	140	57	170	220	130	140
54	<11	<11	<10	<10	<12	<9	<11	<12	<12
60+56	<141	220	220	<122	<145	140	<142	<146	<147
70	150	370	650	120	<55	230	130	110	89
74	71	160	190	57	<34	100	76	71	49
87	100	250	520	46	18	190	35	21	15
95	130	310	530	100	99	190	86	<52	110
90+101	290	690	1500	150	110	540	120	<61	99
99	110	260	490	35	110	330	68	21	14
104	<11	<11	<10	<10	<12	<9	<11	<12	<12
105	110	300	600	33	110	430	69	<12	14
110	260	650	1500	100	31	410	56	37	32
114	<11	11	22	<10	<12	11	<11	<12	<12
118	240	620	1400	89	430	1200	160	19	18
123	20	35	67	18	<12	29	<11	<12	16
132+153	370	860	2000	160	920	1700	270	86	65
138	730	1700	4200	290	1800	5600	740	150	61
141	99	230	590	56	34	290	40	28	28
149	380	900	2000	190	140	840	150	89	140
151	100	250	460	59	66	240	51	32	63
155	<11	<11	<10	<10	<12	<9	<11	<12	<12
156	57	140	400	34	320	430	58	19	<12
157	16	36	92	10	40	74	<11	<12	<12
158	67	140	350	27	42	170	45	20	<12
167	26	74	220	13	51	140	20	<12	<12
170	230	530	1500	130	1100	1800	270	74	19
174	150	360	990	85	34	470	58	43	26
180	300	750	2100	160	1700	2000	300	89	28
183	100	220	590	60	120	360	77	33	33
187	190	440	1200	110	240	1700	420	54	31
188	<11	<11	<10	<10	<12	<9	<11	<12	<12
189	<11	25	76	14	71	65	<11	<12	<12
194	63	160	610	63	760	470	91	42	<12
199	<11	21	74	<10	<12	30	<11	<12	<12
203	64	190	660	62	200	460	110	30	<12
α-HCH	500	900	1100	28	<19	740	<19	<19	<19
β-HCH	170	360	7400	250	470	480	270	200	210
γ-HCH	3500	4700	890	600	<130	1900	1000	620	630
o,p'-DDD	56	140	450	<10	<12	110	48	<12	<12
p,p'-DDD	98	300	1400	61	65	730	140	30	19
o,p'-DDE	24	70	110	15	<160	35	<160	<160	<160
p,p'-DDE	670	1500	4800	410	8800	2900	870	300	45
o,p'-DDT	280	1300	4100	230	<12	720	<11	<12	<12
p,p'-DDT	620	2600	12700	770	260	4300	570	320	120
p,p'-DDT/p,p'-DDE	0.92	1.77	2.66	1.86	0.03	1.48	0.66	1.07	2.77
α/ß-HCH	2.88	2.49	0.14	0.11	n.a.	1.53	n.a.	n.a.	n.a.

Table SI-3c: Norwegian profiles – PBDEs.

	Profile / Layers					
	N-1G			N-2F		
	Oe	Oa	A	Oe	Oa	A
PBDE[1]						
28	<11	<27	<14	<42	<53	<21
47	**61**	<46	<24	**110**	**100**	<35
49	<11	<27	<14	<42	<53	<21
66	<11	<27	<14	<42	<53	<21
85	<11	<27	<14	<42	<53	<21
99	**99**	<150	<81	<230	<290	<120
100	**16**	<27	<14	<42	<53	<21
119	<11	<27	<14	<42	<53	<21
138	<11	<27	<14	<42	<53	<21
153	<11	<27	<14	<42	<53	<21
154	<11	<27	<14	<42	<53	<21
183	<17	<40	<22	<62	<78	<31

[1] Congeners that were < LOD in all samples are not shown.

Table SI-3c continued.

	Profile / Layers						
	N-2G			N-3G			
	Oe	Oa1	Oa2	Oe	Oa	A1	A2
PBDE[1]							
28	<42	<39	<52	<56	<56	<26	<37
47	<71	<66	<88	**450**	**300**	<44	<62
49	<42	<39	<52	**71**	<56	<26	<37
66	<42	<39	<52	**110**	**61**	<26	<37
85	<42	<39	<52	**62**	<56	<26	<37
99	<230	<220	<290	**830**	**480**	<150	<210
100	<42	<39	<52	**100**	**84**	<26	<37
119	<42	<39	<52	<56	<56	<26	<37
138	<42	<39	<52	**320**	<56	<26	<37
153	<42	<39	<52	**270**	<56	**97**	<37
154	<42	<39	<52	<56	<56	<26	<37
183	<62	<58	<78	<83	<83	<39	<55

[1] Congeners that were < LOD in all samples are not shown.

Table SI-3c continued.

	Profile / Layers								
	N-4F				N-4G				
	Oe1	Oe2	Oa	A	Oe	Oa	peat1	peat2	peat3
PBDE[1]									
28	32	35	<26	<24	27	<29	<29	<29	<29
47	600	750	81	<41	1900	650	<49	<50	<49
49	130	100	<26	<24	98	<29	<29	<29	<29
66	140	130	<26	<24	140	<29	<29	<29	<29
85	77	77	<26	<24	74	<29	<29	<29	<29
99	1100	1200	<140	<140	1200	<160	<160	<160	<160
100	120	170	<26	<24	320	82	<29	<30	<29
119	44	39	<26	<24	33	<29	<29	<29	<29
138	170	160	130	140	130	170	<29	<29	<29
153	210	230	32	<24	390	120	<29	51	<29
154	81	75	<26	<24	100	<29	<29	<29	<29
183	370	430	310	<36	370	<43	<44	<44	<43

[1] Congeners that were < LOD in all samples are not shown.

Table SI-4: Congener ratio (R) for vegetation (R-veg), superficial litter (R-litt) and Oi layer (R-Oi).

I-1800	selected PCB congener				
	28+31	**52**	**101**	**138**	**153**
R-veg	1.74	1.86	2.44	1.28	3.44
R-litt	0.19	0.38	0.42	1.53	1.61
R-Oi	0.07	0.13	0.29	1.56	1.71
I-1400	**selected PCB congener**				
	28+31	**52**	**101**	**138**	**153**
R-veg	1.49	1.55	2.64	1.15	2.73
R-litt	0.20	0.37	0.44	1.83	1.78
R-Oi	0.12	0.27	0.39	1.91	1.94
I-1100	**selected PCB congener**				
	28+31	**52**	**101**	**138**	**153**
R-veg	1.15	1.46	2.36	1.69	2.89
R-litt	0.48	0.80	0.87	1.77	2.22
R-Oi	0.41	0.57	0.87	1.73	2.31

Table SI-5a: Estimated fractions (*f*) of PCBs lost from the O horizon by leaching.

$f_{leach\text{-}diss}$ [% year^{-1}]

	I-1800		**I-1400**		**N-1**		**N-2F**		**N-2G**		**N-3**		**N-4F**		**N-4G**	
	min	max	min	max	min	max	min	max	min	max	min	max	min	max	min	max
PCB-28	0.67	1.2	0.31	0.53	1.3	2.6	0.88	1.8	0.43	0.89	0.64	1.3	0.89	1.8	0.66	1.4
PCB-52	0.57	1.0	0.26	0.46	1.1	2.2	0.76	1.6	0.37	0.76	0.55	1.1	0.76	1.6	0.57	1.2
PCB-101	0.40	0.69	0.18	0.32	0.75	1.5	0.52	1.1	0.26	0.53	0.38	0.78	0.53	1.1	0.39	0.81
PCB-138	0.11	0.20	0.05	0.09	0.22	0.44	0.15	0.31	0.07	0.15	0.11	0.22	0.15	0.31	0.11	0.23
PCB-153	0.17	0.30	0.08	0.14	0.33	0.68	0.23	0.48	0.11	0.23	0.17	0.34	0.23	0.48	0.17	0.36
PCB-180	0.04	0.06	0.02	0.03	0.07	0.14	0.05	0.10	0.02	0.05	0.03	0.07	0.05	0.10	0.04	0.07

$f_{leach\text{-}DOC}$ [% year^{-1}]

	I-1800		**I-1400**		**N-1**		**N-2F**		**N-2G**		**N-3**		**N-4F**		**N-4G**	
	min	max	min	max	min	max	min	max	min	max	min	max	min	max	min	max
PCB-28	0.13	0.22	0.05	0.09	0.25	0.52	0.17	0.36	0.09	0.18	0.13	0.26	0.18	0.36	0.13	0.27
PCB-52	0.13	0.22	0.05	0.09	0.25	0.52	0.17	0.36	0.09	0.17	0.13	0.26	0.18	0.36	0.13	0.27
PCB-101	0.13	0.22	0.05	0.09	0.25	0.51	0.17	0.36	0.08	0.17	0.13	0.26	0.17	0.36	0.13	0.27
PCB-138	0.13	0.22	0.05	0.09	0.24	0.50	0.17	0.35	0.08	0.17	0.12	0.25	0.17	0.35	0.13	0.26
PCB-153	0.13	0.22	0.05	0.09	0.25	0.51	0.17	0.35	0.08	0.17	0.12	0.26	0.17	0.35	0.13	0.26
PCB-180	0.12	0.21	0.05	0.09	0.24	0.49	0.17	0.34	0.08	0.17	0.12	0.25	0.17	0.35	0.12	0.26

$f_{leach\text{-}diss} + f_{leach\text{-}DOC}$ [% year^{-1}]

	I-1800		**I-1400**		**N-1**		**N-2F**		**N-2G**		**N-3**		**N-4F**		**N-4G**	
	min	max	min	max	min	max	min	max	min	max	min	max	min	max	min	max
PCB-28	0.80	1.38	0.36	0.62	1.52	3.13	1.06	2.18	0.52	1.06	0.77	1.58	1.07	2.20	0.79	1.63
PCB-52	0.70	1.22	0.32	0.55	1.34	2.76	0.93	1.92	0.45	0.94	0.68	1.39	0.94	1.93	0.70	1.43
PCB-101	0.52	0.91	0.23	0.40	1.00	2.06	0.70	1.43	0.34	0.70	0.51	1.04	0.70	1.45	0.52	1.07
PCB-138	0.24	0.41	0.10	0.18	0.46	0.94	0.32	0.66	0.16	0.32	0.23	0.48	0.32	0.66	0.24	0.49
PCB-153	0.30	0.52	0.13	0.23	0.58	1.19	0.40	0.83	0.20	0.40	0.29	0.60	0.40	0.83	0.30	0.62
PCB-180	0.16	0.28	0.07	0.11	0.31	0.63	0.21	0.44	0.10	0.22	0.16	0.32	0.22	0.45	0.16	0.33

Table SI-5b: Estimated fractions (*f*) of PCBs lost by degradation.

f_{degrad} [% $year^{-1}$] (estimates from ref. 6)								
	I-1800	**I-1400**	**N-1**	**N-2F**	**N-2G**	**N-3**	**N-4F**	**N-4G**
PCB-28	22	22	18	20	20	22	22	22
PCB-52	14	14	11	12	12	14	14	14
PCB-101	2.4	2.4	1.9	2.2	2.2	2.4	2.4	2.4
PCB-153	0.45	0.45	0.36	0.40	0.40	0.45	0.45	0.45
PCB-180	0.25	0.25	0.20	0.22	0.22	0.25	0.25	0.25

Table SI-6: *U* [µg m^{-2}] of selected PCBs and OCPs in the Italian and Norwegian soil profiles.

profile	layer	18	28+31	49	52	101	118	138	153	180	194	α-HCH	γ-HCH	p,p'-DDE	p,p'-DDT
I-1800	Oi	0.05	0.10	0.07	0.19	0.41	0.93	2.2	2.5	1.4	0.22	0.23	0.35	2.7	3.0
	Oe	0.04	0.31	0.42	0.55	2.0	4.5	13	15	10	2.0	0.48	0.86	20	19
	Oa	0.03	0.12	0.13	0.38	1.47	3.5	9.6	11	8.3	1.8	0.28	0.25	19	15
	A1	0.66	0.80	0.12	1.3	2.5	6.8	19	21	15	3.9	0.14	0.69	43	22
	A2	4.5	9.5	4.6	11	4.6	6.3	17	20	13	1.8	0.79	2.8	44	24
	A3	4.1	8.3	2.5	8.5	0.59	1.3	8.4	10	8.7	1.4	1.1	3.0	16	11
	AB	6.0	12	4.9	16	2.0	1.4	4.4	6.3	3.4	1.5	1.2	4.4	7.1	6.3
I-1400	Oi	0.10	0.14	0.16	0.32	0.46	1.05	2.3	2.3	1.2	0.20	0.40	0.48	1.9	3.1
	Oe	0.20	0.54	0.99	1.5	3.3	7.3	18	19	11	2.1	1.8	3.0	16	26
	Oa	0.05	0.21	0.33	0.89	1.7	3.9	9.4	11	7.2	1.8	0.48	0.56	8.8	18
	A1	0.90	1.6	0.24	1.7	3.9	10	33	35	26	8.0	1.6	2.9	45	53
	A2	1.1	2.3	0.34	2.4	1.1	2.3	9.3	12.01	8.0	1.9	0.41	1.6	19	23
	AC	3.3	5.8	0.84	18	0.56	1.2	10.0	13.56	7.5	1.3	1.0	7.1	19	28
I-1100	Oi	0.069	0.134	0.086	0.185	0.282	0.320	0.562	0.750	0.324	0.011	0.095	0.288	0.595	0.914
	A1	8.0	8.3	4.3	20	4.5	8.4	30	33	21	3.4	0.802	5.2	18	19
	A2	6.6	11	5.3	14	4.0	3.1	18	20	15	2.6	1.0	4.1	15	13
	AC	13	18	5.7	25	1.5	3.5	17	20	13	1.4	1.1	7.2	13	10
N-1G	Oe	n.d.	n.d.	0.03	0.06	0.20	0.14	0.59	0.36	0.32	0.04	2.1	1.4	n.d.	0.83
	Oa	0.22	0.36	0.13	0.26	0.44	0.38	1.1	0.57	0.37	0.10	3.8	1.6	1.8	4.1
	A	n.d.	n.d.	n.d.	n.d.	n.d.	0.29	n.d.	n.d.	n.d.	n.d.	11.07	n.d.	0.11	0.15
N-2F	Oe	n.d.	n.d.	n.d.	0.12	0.31	0.24	0.73	0.37	0.29	0.06	1.5	2.4	0.49	0.95
	Oa	n.d.	n.d.	0.28	0.46	1.6	1.6	4.9	2.6	2.4	0.43	1.8	n.d.	1.1	2.1
	A	1.5	7.2	0.89	1.1	0.57	0.34	0.95	0.60	0.36	0.07	3.4	5.8	0.75	1.1
N-2G	Oe	n.d.	n.d.	n.d.	n.d.	n.d.	0.15	0.37	0.20	0.12	n.d.	1.3	2.8	0.33	0.67
	Oa1	n.d.	n.d.	0.14	0.31	0.94	0.90	2.5	1.2	0.88	0.24	0.25	1.5	2.9	6.2
	Oa2	1.3	4.7	0.4	0.49	n.d.	0.20	0.55	0.30	0.24	0.09	n.d.	1.9	0.70	1.1
N-3G	Oe	n.d.	n.d.	n.d.	n.d.	0.49	0.90	2.5	1.2	0.91	0.22	0.55	1.9	2.1	2.3
	Oa	0.51	n.d.	0.31	0.23	1.1	1.8	5.1	2.3	2.0	0.52	0.93	2.9	2.5	5.2
	A	1.7	9.7	0.95	1.1	n.d.	0.17	0.76	0.43	0.35	0.16	0.76	1.2	0.89	0.48
	A	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.24	n.d.	n.d.	n.d.
N-4F	Oe1	n.d.	n.d.	0.08	0.08	0.44	0.38	1.1	0.57	0.46	0.10	0.77	5.4	1.0	0.95
	Oe2	n.d.	n.d.	0.19	0.20	1.1	0.95	2.6	1.3	1.1	0.25	1.4	7.3	2.3	4.0
	Oa	n.d.	n.d.	0.31	0.41	2.4	2.4	6.9	3.3	3.5	1.0	1.7	1.4	7.8	21
	A	0.31	1.5	0.20	0.27	0.28	0.17	0.54	0.30	0.30	0.12	0.05	1.1	0.77	1.4
N-4G	Oe	0.38	n.d.	0.10	0.18	0.34	1.3	5.6	2.8	5.4	2.3	n.d.	0.95	27	0.80
	Oa	n.d.	n.d.	0.17	0.53	1.7	3.8	18	5.6	6.3	1.5	2.3	5.9	9.3	14
	peat1	1.7	5.0	0.52	0.70	0.38	0.49	2.3	0.85	0.95	0.29	n.d.	3.2	2.7	1.8
	peat2	0.64	2.7	0.30	0.38	n.d.	0.05	0.43	0.25	0.25	0.12	n.d.	1.8	0.86	0.92
	peat3	0.96	2.6	0.28	0.39	0.28	0.05	0.17	0.18	0.08	n.d.	n.d.	1.8	0.13	0.35

n.d. not detected

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