SUPPLEMENTARY MATERIALS:

SPATIAL AND TEMPORAL TRENDS OF ORGANIC POLLUTANTS IN VEGETATION FROM REMOTE AND RURAL AREAS

Short title: Global distribution of POPs in vegetation

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Table S1. Classification of the persistent organic pollutants (POPs) and polycyclic aromatic hydrocarbons (PAHs) in this study. Phen –Phenanthrene, Ant – Anthracene, Ftn – Fluoranthene, Pyr – Pyrene, BaA – Benz[a]anthracene, Ch – Chrysene, BbF – Benzo[b]fluoranthene, BkF – Benzo[k] fluoranthene, B[a]P – Benzo[a]pyrene.

Use		**POP**	**LogK_{OA}**	**LogK_{OW}**
Intentional	Fungicide and combustion byproduct of chlorinated organics	HCB	7.21	5.64
	Insecticides	HCHs		
		α-HCH	7.46	3.94
		γ-HCH	7.74	3.83
	Insecticides	DDTs		
		p,p'-DDE	9.70	4.20
		p-p'-DDT	9.73	6.39
	Industrial, dielectric/hydraulic fluid	PCBs		
		PCB28		5.71
		PCB52	7.82	5.79
		PCB101	8.24	6.40
		PCB118	9.82	6.57
		PCB138	9.06	6.70
		PCB153	9.55	6.80
		PCB180	10.20	7.21
	Industrial, flame retardant	PBDEs		
		BDE47	10.53	6.55
		BDE99	11.31	7.13
		BDE183	11.96	8.10
		BDE209	15.27	9.97
Unintentional	Forest fires and geochemical processes	PAHs		
		Phen	7.68	4.57
		Ant	7.71	4.45
		Ftn	8.76	5.22
		Pyr	8.93	5.18
		BaA	10.28	5.79
		Ch	10.3	5.73
		BbF	11.34	6.11
		BkF	11.37	6.11
		BaP	11.56	6.13

Sources: Harner and Shoeib [1], Tittlemier, et al. [2], Wania and Dugani [3], Xiao, et al. [4], Veltman, et al. [5], Mackay, et al. [6], Odabasi, et al. [7], and Daly, et al. [8], Finizio, et al. [9], Makino [10], Shiu and Ma [11]

Table S2. Plant types, common names, scientific names (when available), and POP concentrations (mean ± standard deviation in $ng \cdot g^{-1}$ dry weight) from the data presented in this study.

Group	Common name	Species	N	HCB	∑HCHs	∑DDTs	∑PCBs	∑PBDEs	∑PAHs
Lichen (plant-like composite organism)	lichen	*Lecanora aspidophora*, *Stereocaulon glabrum*, *Umbilicaria* spp., *Umbilicaria propagulifera*, *Usnea sulphurea*, *Hypogymnia physodes*, *Parmotrema hypoleucinum*, *Usnea* spp., *Usnea antarctica*, *Usnea aurantiaco-atra*	59	2.59 ± 3.36	42.73 ± 56.57	6.33 ± 7.98	28.86 ± 33.04	0.17 ± 0.06	641.7 ± 772.1
Bryophyta	moss	*Andreaea regularis*, *Bryum algens*, *Drepanocladus uncinatus*, *Brachytecyum* sp., *Hylocomium splendens*, *Pleurozium schreberi*, *Hypnum cupressiforme*, *H. plumaeformae*, *Isopterygium minutirameum*, *Sanionia uncinata*, *Sphagnum* spp., *Syntrichia princeps*	213	1.97 ± 3.92	15.41 ± 24.71	2.73 ± 3.75	19.80 ± 20.38	1.23 ± 1.41	761.2 ± 847.8
Lichen & moss			15	0.41 ± 0.39	10.74 ± 13.87	9.52 ± 16.46			
Gymnosperm	cedar	*Cedrus deodara*, *Juniperus virginiana*	30	1.16 ± 1.59	0.27 ± 0.23	2.88 ± 2.83	15.90 ± 12.68	2.79 ± 2.21	
	cypress	*Cupressus torulosa*	4	1.03 ± 0.02	3.34 ± 0.92	11.17 ± 0.96			
	larch	*Larix decidua*	5	0.61	0.34		2.18 ± 1.25		
	pine	*Pinus canariensis*, *P. densata*, *P. densiflora*, *P. halepensis*, *P. koraiensis*, *P. massoniana*, *P. tabulaeformis*, *P. maximartinezii*, *P. nigra*, *P. pinaster*, *P. pinea*, *P. strobus*, *P. sylvestris* (N = 388), *P. tabulaeformis*, *P. taeda*, *P. thunbergii*, *P. uncinata*, *P. virginiana*	727	1.82 ± 1.52	14.58 ± 15.94	2.72 ± 8.38	19.65 ± 32.91	267.7 ± 1035.3	542.3 ± 1120.8
	spruce	*Picea abies*, *Tsuga dumosa*	47	0.89 ± 0.50	3.03 ± 2.93	7.93 ± 11.91	31.58 ± 164.9	2.11	97.42 ± 123.3
Angiosperm	ash	*Fraxinus excelsior*, *Sorbus aucuparia*	13				1.71 ± 0.69		242.10
	beech	*Fagus sylvatica*	6	0.19	0.36		1.84 ± 1.17		
	camphor tree	*Cinnamomum camphora*	49					19.32 ± 57.24	
		Cinnamomum camphora, *Buxus* sp.	3					29.79 ± 24.31	
	chestnut	*Castanea sativa*	5	0.21	0.24		1.98 ± 0.94		
	elm	*Ulmus* spp.	3					57.45 ± 83.47	
	eucalyptus	*Eucalyptus* spp.	1					33.83	
	grass	*Deschampsia antarctica*, Poaceae	69	0.14 ± 0.08		0.08 ± 0.03	0.33 ± 0.30	1.55 ± 1.95	7.72 ± 2.86
	hazelnut	*Corylus avellana*	7	0.04	0.09		1.56 ± 0.72		577.5
	lime	*Tilia cordata*	4				1.71 ± 0.94		
	mango	*Mangifera indica*	10	0.11 ± 0.01	13.52 ± 37.58	24.30 ± 30.24			
	maple	*Acer pseudoplatanus*, *A. saccarum*	39	0.06	0.21		1.46 ± 0.90		766.7 ± 758.1

oak	*Quercus* spp., *Q. mongolica*, *Q. robur*	19					571.1 ± 661.8	283.20
poplar	*Populus*	1					101.5	
olive	*Olea europaea*	7	31 ± 16	91 ± 52	81 ± 63	255 ± 166		
corn	*Zea mays*	30						26.52 ± 11.85
oak, cherry tree, hornbeam, alderwood ashwood		1	0.33	81.20	103.90			
pearlwort	*Colobanthus quitensis*	3	0.53 ± 0.69	1.05	0.08 ± 0.04	7.31 ± 10.17	0.28	9.52

Sources: Lichen [12-21]; Bryophyta [13,15,17-19,21-39]; Lichen and moss [16]; Cedar [40,41]; Cypress [42]; Larch [43,44]; Pine [12,19,20,27,28,41,42,45-79]; Spruce [42-44,57,80-84]; Ash [43,85]; Beech [43,44]; Camphor tree [86,87]; Chestnut [43,44]; Elm [79]; Eucalyptus [68]; Grass [15,88,89]; Hazelnut [43,85]; Lime [43]; Mango [16]; Maple [43,44,72]; Oak [49,79,85]; Olive [90]; Poplar [79]; Corn [72]; Oak, cherry tree, hornbeam, alderwood and ashwood [36]; Pearlwort [15,18,21]

Table S3. Multilevel model of the effects of abiotic and biotic variables on the concentrations of POPs in plants. Constant and varying effects by congeners are included for the predictor variables included in the lowest-AIC models. All models accounted for spatial correlation structure and nugget (which were significant in all cases, $P < 0.001$) and were run using the restricted maximum likelihood criterion for optimizing parameter estimates. MAT, mean annual temperature; MAP, mean annual precipitation. •$P < 0.1$, *$P < 0.05$, **$P < 0.01$, ***$P < 0.001$

	HCB (N = 419)	HCHs (N = 830)		DDTs (N = 754)	PCBs (N = 72)		PBDEs (N = 188)		PAHs (N = 648)	
	Fixed	Fixed	Random	Fixed	Fixed	Random	Fixed	Random	Fixed	Random
Year	-0.35***	-0.70***		0.44	-0.28***	0.05*			0.24***	
Latitude					-0.17***					
Altitude					0.15**					
MAT		0.18*		-0.10*	0.02		0.28*		0.43***	
MAP					-0.11***					
Group, lichen	0.44*								-0.19	0.33***
Group, Bryophyta	0.17				0.98***				-0.75*	0.25***
Group, gymnosperm	0.39**				-0.04				-0.75*	0.48***
Group, angiosperm										
Continent, North America						0.42***		0.74***		
Continent, Central & South America			0.43***							
Continent, Europe	0.70***	1.14*	0.02***		1.26***	0.13***	-0.21	0.54***		
Continent, Africa	0.06	-0.25	0.15***							
Continent, Asia	0.96***	1.97***	0.19***				-0.58	0.61***		
Land use, rural	-0.89***	0.14*		-1.21**	0.03				0.18*	
Year:rural	-0.59***	0.25***		-1.21***						
MAT:rural				0.40***	0.62***				-0.37***	

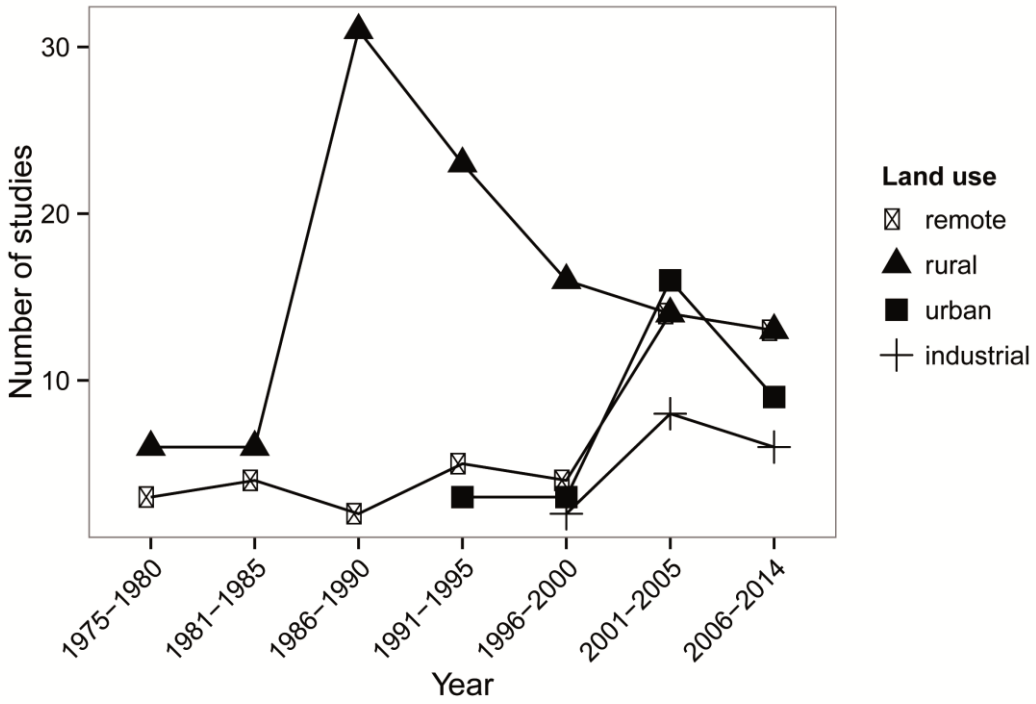

Fig. S1. Temporal evolution of the number of studies on POPs and PAHs in vegetation at remote, rural, urban, and industrial sites.

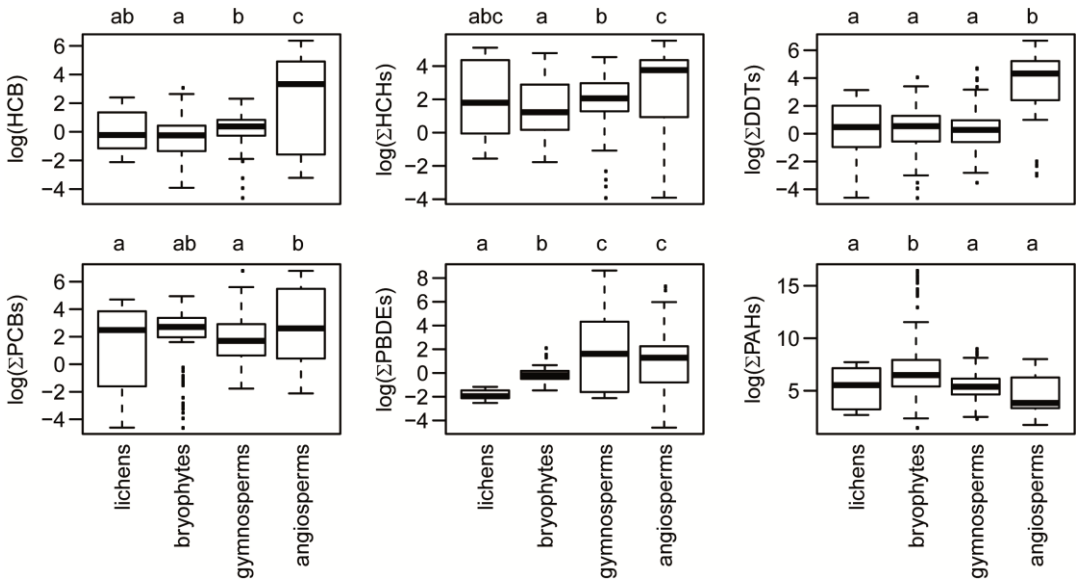

Fig. S2. Variability of the logarithm of the concentration of POPs and PAHs in each vegetation group. Different letters indicate significant differences among groups (Tukey's HSD, $P < 0.05$). Interquartile ranges (25th and 75th percentile) are shown by the height of the boxes, and the horizontal lines represent medians (50th percentile). Whiskers range from the 10th to 90th percentiles, and values outside this range are indicated by small squares. Concentrations are in $ng \cdot g^{-1}$ dry weight.

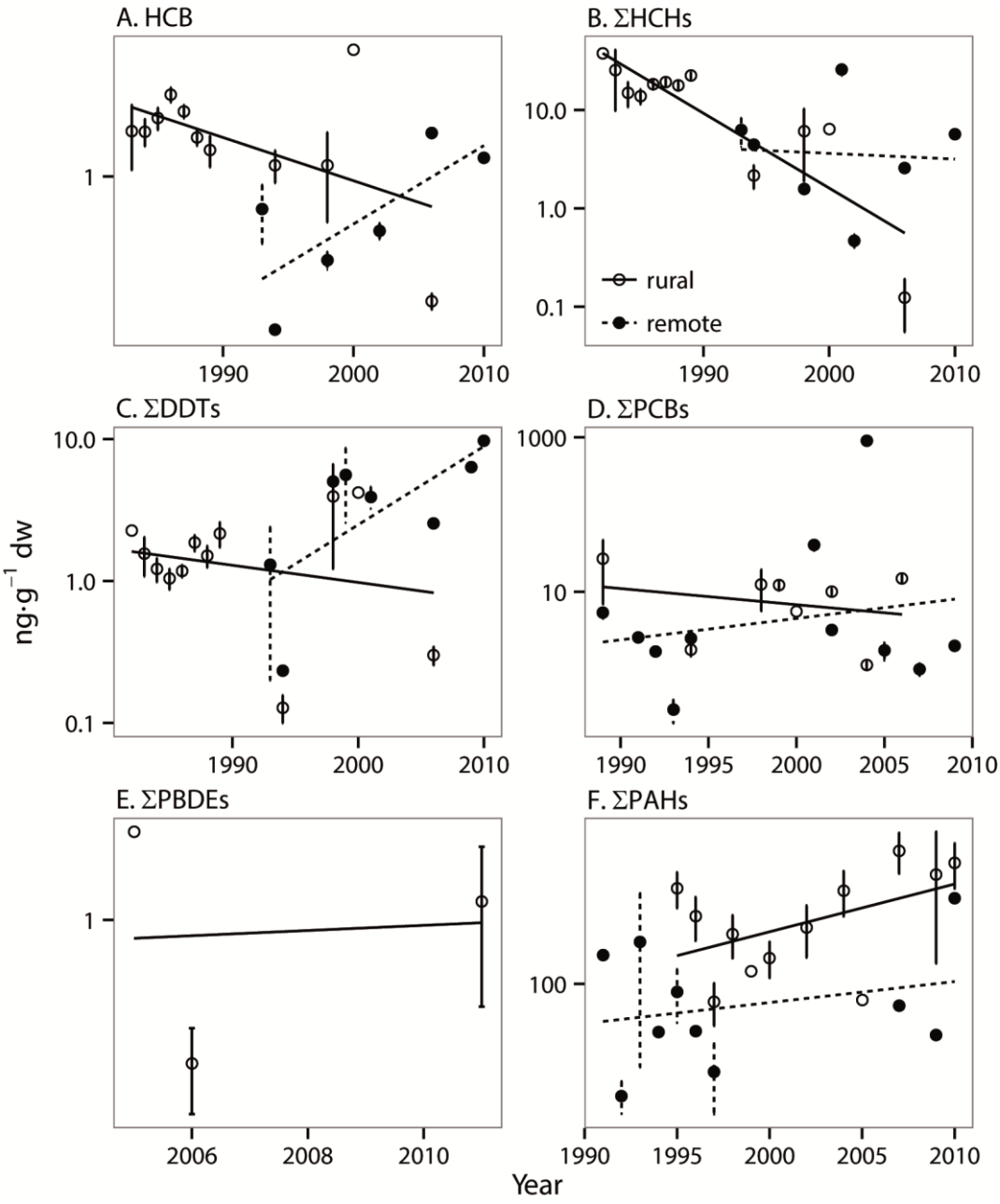

Fig. S3. Individual relationships among HCB, ΣHCHs, ΣDDTs, ΣPCBs, ΣPBDEs, and ΣPAHs (ng·g^{-1} dry weight) and year in gymnosperms. The equations for remote and rural areas are: A) HCB rural: y = -0.04x + 74, r^2 = 0.12, df = 1,303, P < 0.0001, and HCB remote: y = 0.08x - 152, r^2 = 0.56, df = 1,84, P < 0.0001; B) ΣHCHs rural: y = -0.08x + 171, r^2 = 0.33, df = 1,306, P < 0.0001, and ΣHCHs remote: y = -0.02x + 32, r^2 = 0.02, df = 1,101, P = 0.1; C) ΣDDTs rural: y = -0.02x + 40, r^2 = 0.02, df = 1,306, P = 0.009, and ΣDDTs rural: y = 0.03x - 58, r^2 = 0.10, df = 1,104, P < 0.001; D) ΣPCBs rural: y = 0.06x - 113, r^2 = 0.19, df = 1,77, P < 0.0001, and ΣPCBs remote: y = 0.02x - 31, r^2 = 0.02, df = 1,56, P = 0.3; E) ΣPBDEs rural: y = 0.05x - 98, r^2 = 0.06, df = 1,8, P = 0.5; and F) ΣPAHs rural: y = 0.03x - 55, r^2 = 0.11, df = 1,68, P = 0.005, and ΣPAHs remote: y = 0.02x - 38, r^2 = 0.14, df = 1,15, P = 0.1.

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