Supplementary Information

Estimating Overall Persistence and Long-Range Transport Potential of Persistent Organic Pollutants: A Comparison of Seven Multimedia Mass Balance Models and Atmospheric Transport Models

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1 Chemical Property Data for the 14 Reference Chemicals

Table S1: Physical-chemical properties and degradation rate constants of the 14 chemicals selected for the P_{ov} and LRTP rankings.

Chemical	Vapor pressure Pa (25ºC)	Solubility mol.m^{-3} (25ºC)	K_{aw} –	K_{ow} –	k_{air} s^{-1}	k_{soil} s^{-1}	k_{water} s^{-1}	k_{sed} s^{-1}	k_{veg} s^{-1}
aldrin	6.37E-2[a]	2.63E-3[a]	9.77E-3[a]	1.78E+6[a]	6.64E-5[o]	5.03E-8[o,e]	7.20E-8[o]	7.20E-9[o]	6.64E-5[v]
atrazine	3.87E-5[b]	1.42E-1[b]	1.10E-7[b]	5.62E+2[b]	3.11E-5[h]	2.36E-7[p]	2.33E-7[h]	1.91E-8[h]	3.11E-5[v]
B[a]P	6.32E-6[a]	1.14E-4[a]	2.24E-5[a]	8.92E+5[a]	3.63E-5[u,j]	1.13E-8[q]	1.13E-7[q]	3.50E-9[u]	3.63E-5[v]
BDE-47	2.15E-4[c]	1.94E-4[c]	4.46E-4[c]	2.43E+6[c]	7.52E-7[k]	9.63E-9[k]	9.63E-9[k]	9.63E-10[k]	7.52E-7[v]
BDE-99	3.63E-5[c]	6.86E-5[c]	2.14E-4[c]	5.72E+6[c]	4.12E-7[k]	9.63E-9[k]	9.63E-9[k]	9.63E-10[k]	4.12E-7[v]
biphenyl	4.21[a]	1.66E-1[a]	1.05E-2[a]	1.10E+4[a]	4.86E-6[l]	3.50E-7[r]	1.13E-6[r]	1.13E-7[l]	4.86E-6[v]
CCl4	1.65E+4[d]	4.64[d]	1.44[f]	6.76E+2[d]	2.90E-10[m]	3.15E-8[o]	3.15E-8[o]	1.60E-7[m]	2.90E-10[v]
HCB	1.01E-1[a]	1.32E-3[a]	3.09E-2[a]	4.07E+5[a]	1.96E-8[n]	5.64E-9[o]	5.64E-9[o]	5.64E-10[n]	1.96E-8[v]
HCBD	1.78E+1[d]	1.28E-2[d]	5.59E-1[d]	2.42E+4[d]	1.62E-8[u,j]	4.58E-8[d,s]	8.02E-8[t]	8.02E-9[u]	1.62E-8[v]
PCB-153	5.51E-4[a]	3.02E-5[a]	7.41E-3[a]	7.24E+6[a]	1.16E-7[j]	3.50E-10[j]	3.50E-9[j]	1.13E-9[j]	1.16E-7[v]
PCB-180	1.32E-4[a]	1.70E-5[a]	3.09E-3[a]	1.41E+7[a]	7.25E-8[j]	1.93E-10[j]	3.50E-9[j]	1.13E-9[j]	7.25E-8[v]
PCB-28	2.58E-2[a]	8.91E-4[a]	1.17E-2[a]	4.57E+5[a]	7.55E-7[j]	1.93E-8[j]	3.50E-8[j]	1.13E-8[j]	7.55E-7[v]
p-cresol	1.51E+1[e]	1.10E+2[e]	5.51E-5[e]	9.33E+1[e]	3.21E-5[e]	4.81E-5[e]	3.57E-5[e]	3.57E-6[e]	3.21E-5[v]
α-HCH	2.26E-1[a]	3.55E-1[a]	2.57E-4[a]	7.59E+3[a]	1.02E-7[n]	5.94E-8[o]	5.94E-8[o]	3.50E-9[n]	1.02E-7[v]

[a] Schenker et al., 2005, [b] US EPA, 2005, [c] Wania and Dugani, 2003, [d] Mackay et al., 1992, [e] Mackay et al., 1999, [f] Hunter-Smith et al., 1983, [h] Fenner et al,. 2003, [j] Wania and Daly, 2002, [k] Gouin and Harner, 2003, [l] Anderson and Hites, 1996, [m] Atkinson et al., 1989, [n] Brubaker and Hites, 1998, [o] Howard, 1991, [p] Lanz, 2005, [q] Mackay et al., 1992a, [r] Mackay et al, 1992b, [s] Eisenberg and McKone, 1998, [t] HSDB, 2001, [u] Howard and Meylan, 1997, [v] degradation rate constants in vegetation assumed equal to those of air

2 Description of the Seven Models

EVn-BETR

The European Variant Berkeley–Trent model (EVn-BETR) is a fugacity-based model (Mackay, 2001) that comprises 50 regions, with 4 regions in the periphery to describe the world outside Europe. Each region represents an area of approximately 500 km x 500 km (5ºx5º), with the whole model domain covering an area from 38.7 ºN to 61.1 ºN latitude and 10.1ºW to 39.4 ºE longitude (Prevedouros et al., 2004). Each region consists of seven environmental compartments: lower (0–1000 m) and upper air (1000–2000 m), soil, vegetation, ocean water, fresh water and sediment. Detailed information on the model construction can be found in MacLeod et al. (2001) and Woodfine et al. (2001). In Evn-BETR, the long-range transport potential of chemicals is calculated as the average distance from a point source at which the chemical's concentration has dropped to 38% (1/*e*) of its initial concentration. The average distance is calculated using the predicted air concentrations in the eight boxes surrounding the source box.

The atmospheric transport between EVn BETR grid cells has been derived from historical data derived from the European Centre for Medium-Range Weather Forecasts (ECMWF) global circulation model. Forward trajectory data were calculated using 6-hourly operational analyses of the three components of the wind and surface pressure, which were then interpolated on to a 1.5° by 1.5° grid. The output data from the trajectories consisted of latitude, longitude and pressure of the trajectory every 30 min. Air was allowed to move freely to all other regions within the two atmospheric heights of 500 and 2000 m above sea level. The trajectory data were then used to produce an average wind rose for all the model segments for the period of study (1997–2001). The wind roses were then converted into a connectivity flux matrix for each atmospheric height, using a matrix technique described by Woodfine et al. (2001).

MSCE-POP

MSCE-POP is a multi-compartment atmospheric transport model, describing processes in and exchange between four environmental compartments (atmosphere, soil, sea water, and vegetation). Except the vegetation compartment, all compartments are vertically segmented into a number of layers. There are regional and hemispherical versions of the model. The spatial resolution of the latter version, which was used in the EMEP POP model inter-comparison study, is 2.5 x2.5° and the model domain covers the total Northern Hemisphere. Lateral transport of compounds by air and by seawater is taken into account in the model. (Malanichev et al., 2004, Gusev et al., 2005).

ClimoChem

ClimoChem is a multi-compartment mass balance box model that covers the entire global system. Compartments included are soil, oceanic surface water, troposphere air, vegetation and vegetation soil. ClimoChem consists of a flexible number (typically 10 to 30) of latitudinal zones with different temperatures and compartment volumes. ClimoChem does not have a spatial resolution in the East-West direction; in North-South direction, the spatial resolution is given by the number of zones, n, and the width of a zone is equal to $180/n$ degrees latitude (Scheringer et al., 2000). The transport mechanism implemented in the model is large-scale eddy diffusion in atmosphere and oceans. Eddy diffusion coefficients have been derived from tracer measurements in atmosphere and ocean water (Keeling and Heimann, 1986; Okubo, 1971). For the atmosphere, these eddy diffusion coefficients represent the long-term average of the intra- and interhemispheric mixing. For the ocean, they represent the growth of a "patch" of contaminant that is caused by turbulent mixing caused by large-scale eddies. In the ClimoChem model, they describe the exchange of air and water between adjacent latitudinal zones.

SimpleBox

SimpleBox is a nested level III and level IV 'Mackay type' multimedia fate model consisting of ten environmental compartments on local, regional, continental and global scales (Brandes et al., 1996; Den Hollander et al., 2004). The regional and continental scales distinguish an air compartment (atmospheric mixing layer), a sea water compartment with a sediment compartment, a fresh water compartment with a sediment compartment, and three types of soil compartments (natural, agricultural, other soil) with corresponding vegetation compartments. SimpleBox is a generic model, in which the default settings are set to match the European Union procedures for the evaluation of substances (Brandes et al., 1996). Advective transport (e.g. wet and dry deposition, wind flow, water transport, sedimentation), as well as diffusive transport (e.g. gaseous deposition, volatilization) between the different environmental compartments and spatial scales is accounted for in the model. Since SimpleBox is a non-spatial model, transport distances of chemicals are not calculated directly. The relative long-range transport potential of a compound can be estimated based on the fraction of the amount released that is exported across the boundaries of the model: for chemicals with high LRT, this fractions is close to one, for chemicals with low LRT, this fraction is close to zero.

ADEPT

ADEPT (Atmospheric DEPosition and Transport model for risk assessment) is a diagnostic model that calculates concentrations in air and deposition fluxes to the underlying surface; Roemer et al., 2004). The model is derived from the LOTOS-EUROS model (Schaap et al., 2005). It covers the European continent (30°N-70°N; and 10°W to 60°E), but it can be adjusted to sub-domains. There is no exchange with water, soil and vegetation, only a flux (loss term) to the underlying surface. Transport characteristics are taken from the LOTOS-EUROS model by means of source receptor matrices for inert species. Loss by chemistry, wet and dry deposition is modeled on the basis of the average transport time from source to receptor, and by means of average values for OH, O3, photolysis, atmospheric and surface resistances, precipitation and Henry coefficients.

G-CIEMS

G-CIEMS is a GIS-based geo-referenced multimedia fate model consisting of environmental compartments of gridded air cells, polygon and line-based surface catchments and river structures, and polygon-based sea segments (Suzuki et al., 2005). The model is now mainly applied to the Japan-regional environment with detailed (ca. 5 km) resolutions. As the model is flexible to any geographical conditions within the computational limitations, a generic simple framework consisting of only several boxes including the specified European modeling domain and surrounding areas was used in this study, so that essential comparison of the multimedia fate processes would be possible.

3 Statistics of P_{ov} and LRTP Rankings of the 14 Reference Chemicals

Table S2: average deviations of P_{ov} rankings from average ranking of the 14 chemicals: average over all 14 chemicals in each of the 6 models (left) and average over the 6 models for each chemical (right).

model	average deviation of all 14 chemicals	chemical	average deviation of all 6 models
CliMoChem	0.428	p-cresol	0.333
MSCE-POP	0.428	CCl4	0.333
G-CIEMS	0.428	a-HCH	0.50
OECD Tool	0.571	BDE-47	0.50
SimpleBox	1.0	aldrin	0.667
EVN-BETR	1.29	biphenyl	0.667
		BDE-99	0.667
		HCB	0.667
		PCB-180	0.667
		atrazine	0.833
		BaP	0.833
		PCB-28	0.833
		PCB-153	0.833
		HCBD	1.33
		average of all 14 chemicals	0.69

Table S3: average deviations of LRTP rankings from average ranking of the 14 chemicals: average over all 14 chemicals in each of the 8 models (left) and average over the 8 models for each chemical (right).

model	average deviation of all 14 chemicals	chemical	average deviation of all 8 models
OECD Tool (CTD)	0.714	p-cresol	0.375
OECD Tool (TE)	1.0	aldrin	0.5
G-CIEMS	1.29	BaP	0.625
MSCE-POP	1.29	BDE-47	1.125
SimpleBox	1.29	HCB	1.375
EVN-BETR	1.43	CCl4	1.375
ADEPT	2.0	PCB-153	1.375
CliMoChem	2.43	HCBD	1.5
		biphenyl	1.75
		atrazine	1.75
		a-HCH	1.875
		PCB-28	1.875
		PCB-180	2.25
		BDE-99	2.25
		average of all 14 chemicals	1.43

4 Mass Balance Estimates for PCB-153 in the Year 2000

	MSCE-POP		CliMoChem		SimpleBox		EVn-BETR		G-CIEMS	
Volume										
air (m3)	1.26E+17		6.19E+16		9.59E+15		1.06E+16		1.05E+17	
water (m3)	9.83E+14		8.41E+14		8.68E+14		4.25E+14		9.84E+14	
sediment (m3)	na				1.35E+11		3.81E+09		2.18E+11	
soil (m3)	1.11E+12		5.92E+11		6.11E+11		5.58E+11		5.57E+11	
vegetation (m3)	nr		5.00E+09		8.34E+09		3.26E+09		6.25E+08	
Concentration 2000										
air (pg/m3)			0.44		3.7		7.5		4.9	
interface with ocean	3.7									
interface with soil	7.4									
interface with vegetation	7.4									
water (pg/L)			0.21		2.0		1.70		0.2	
interface with atmosphere	0.18									
sediment (pg/g)	na		na		107		73.0		2	
soil (pg/g)			12.07		40		26.0		63	
interface with atmosphere	168									
vegetation (pg/g)	2634		16.24		18		105		1363	
Mass 2000										
air (kg)	107	0.1%	27	0.1%	38	0.1%	79	0.2%	61	0.1%
water (kg)	1285	1.4%	180	1.0%	1759	3.6%	722.9	2.0%	171	0.4%
sediment (kg)	na				20187		667.5	1.8%	717	1.7%
soil (kg)	84167	95%	17860	97.8%	46687	96%	34804	95.1%	41809	99%
forest litter (kg)	2709							0.0%		
vegetation (kg)	684	0.8%	203	1.1%	161	0.3%	342	0.9%	42	0.1%
total mass (kg)	88952	100%	18270	100.0%	48645	100%	36616	100.0%	42082	100%
Changes in inventory from 1999 to 2000										
air (kg/yr)	4.4	4.1%	-2.28	-8%	-2.8	-7.5%	-7	-8.8%	-29.7	-48.9%
water (kg/yr)	-78	-6.1%	-12.60	-7%	-158	-9.0%	-62	-8.6%	-108	-63.2%
sediment (kg/yr)					-1862	-9.2%	-22		642	89.6%
soil (kg/yr)	-3782	-4.5%	-412	-2%	-3251	-7.0%	-550	-1.6%	-3390	-8.1%
forest litter (kg/yr)	-188	-7.0%								
vegetation (kg/yr)	-25	-3.6%	-18	-9%	-13	-8.2%	-27	-7.9%	-46	-112.0%
total (kg/yr)	-4069		-445		-5288		-668		-2932	
Mass balances for individual media										
air (kg/yr)	-35	-32.7%	1	2.9%	-9	-24.3%	10.0	12.6%	-187	-308.7%
water (kg/yr)	-46	-3.6%	0	-0.3%	51	2.9%	-4.0	-0.6%	-248	-144.5%
sediment (kg/yr)					101	0.5%	94.0	14.1%	582	81.2%
soil (kg/yr)	381	0.5%	-35	-0.2%	-997	-2.1%	-501.0	-1.4%	-4206	-10.1%
forest litter (kg/yr)	-381	-14.1%								
vegetation (kg/yr)	0	0.0%	5	2.3%	-9	-5.6%	365.0	106.6%	-90	-216.9%
total (kg/yr)	-80	-0.1%	-30	-0.2%	-864	-1.8%	-36.0	-0.1%	-4149	-9.9%
Mass budget										
emission to model domain (kg/yr)	5546	100%	995	100%	5535	100%	7568	100%	5452	100%
emission to rest of N hemishere (kg/yr)	5844								5446	
net export from domain with air (kg/yr)	3474	63%	208	21%	676	12%	2797	37%	2832	52%
net export from domain with water (kg/yr)	-17.2	-0.3%	8.2	0.8%	1251	23%	3041.00	40%	412	7.6%
reaction in air (kg/yr)	45	0.8%	12	1.2%	41.7	0.8%	87.0	1.1%	89.8	1.6%
reaction in water (kg/yr)	33	0.6%	9.2	0.9%	447	8.1%	81.00	1.1%	54	1.0%
reaction in sediment (kg/yr)					2276	41%	43.00	0.6%	54	1.0%
reaction in soil (kg/yr)	4343	78%	844	85%	5238	95%	1861.00	25%	4897	90%
reaction in litter layer (kg/yr)	569	10%								
reaction in vegetation (kg/yr)	0	0%	101	10%	180	3.2%	110.0	1.5%	161	3.0%
burial in sediment (kg/yr)					75	1.4%			0.04	0.0%
leaching from soil (kg/yr)					0.44	0.0%			0.00	0.0%
removal of plants (kg/yr)					112	2.0%			0	0.0%
deposition to deep sea (kg/yr)	1213	22%	289	29%						
removal from model domain (kg/yr)	9660	174%	1471	148%	10298	186%	8020.00	106%	8501	156%
Intermedia flow										
dry deposition to water (kg/yr)	58				327				304	
wet deposition to water (kg/yr)	461				1457				525	
gas absorption to water (kg/yr)	620				378				560	
total deposition to water	1140	21%	258	26%	2162	39%	3171	42%	1389	25%
dry deposition to soil (kg/yr)	1006				355				189	
wet deposition to soil (kg/yr)	1530				1647				326	
gas absorption to soil (kg/yr)	-2864				213				1	
total deposition to soil	-329	-5.9%	215	22%	2215	40%	810	11%	517	9.5%
dry deposition to vegetation (kg/yr)	246				28				156	
wet deposition to vegetation (kg/yr)	0				40				270	
gas absorption to vegetation (kg/yr)	1001				384				416	
total deposition to vegetation	1247	22%	304	31%	453	8.2%	700	9.2%	841	15%
total atmospheric deposition	2057	37%	777	78%	4830	87%	4681	62%	2747	50%
sedimentation (kg/yr)					590	11%	115.000	1.5%	1279	23%
run off to surface water (kg/yr)			35	3.55%	18	0.3%				0.0%
litter fall to soil (kg/yr)	1271	23%	217	22%	183	3.3%	252.0	3.3%	817	15.0%

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