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# **Residue of DDT and HCH in Fish from Lakes and Rivers in the World**

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Additional information is available at the end of the chapter

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## **1. Introduction**

Organochlorine pesticides such as DDT and HCH had been abundantly produced and used since the around twentieth century in all over the world and environmental pollution, food contamination and human body pollution had developed into social problem because of their high bioconcentration potential and high persistency. The use and manufacturing of DDT and HCH were forbidden in many countries including Japan in all over the world in the first half in 1970's.

In Japan, the use of organochlorine pesticides registered in persistent organic pollutants (POPs) was prohibited, for example, in 1971 for DDT and HCH and in 1986 for chlordane. Levels of DDT and HCH have been monitored annually in wildlife such as fish, shellfish and bird from a freshwater lake, 17 sea areas and 2 land areas in Japan since 1979 [1-31]. Lake Biwa, the largest in Japan, was selected as a freshwater lake and a freshwater fish, Japanese dace from the lake was selected as a wildlife sample.

In the world, a few reports have been published for long-term monitoring of POPs in aquatic biota from lakes only in USA and Sweden. In USA, the use of DDT was prohibited in 1972. Levels of POPs such as DDT, chlordanes, Mirex, Dieldrin have been monitored annually in fish from the Grate Lakes since 1970 [32, 33]. In Sweden, the use of DDT was prohibited in 1970. Levels of DDT, HCB and HCH have been monitored annually in fish from Lake Storvindeln since 1968 [34].

On the other hand, many field data have been published for temporal monitoring of DDT and HCH in fish from lakes [35-70] and rivers [40, 51, 68, 71-101].

This study was performed for the accumulation of fundamental data on DDT and HCH contamination of fish in lakes and rivers in the world to evaluate their concentration changes by POPs Regulation. The data were collected from the published reports in which

the accuracy in the chemical analyses of the pesticides was over the standard level. This chapter consisted of (1) Residue of T-DDT and T-HCH in fish from lakes and rivers in the world, (2) Long-term trends of T-DDT and T-HCH in fish from lakes in the world, (3) Composition of T-DDT and T-HCH in fish from lakes and rivers in the world.

1. Each of the T-DDT and T-HCH concentration data in fish from lakes and rivers was compared for the 38 lakes surveyed in 8 countries of Europe and America and 8 countries of Asia and Africa from 1995 to 2008 and for the 28 rivers surveyed in 8 countries of Europe and America, 4 countries of Asia, Africa and Oceania from 2000 to 2009.
2. Long-term trends of T-DDT and T-HCH in Japanese dace from Lake Biwa were shown from 1979 to 2009 and half-lives ( $t_{1/2}$ ) were calculated for T-DDT and T-HCH. The  $t_{1/2}$  values were 9 years for T-DDT and 4 years for T-HCH. Similarly, long-term trends of T-DDT in fish from Lake Biwa, Lake Ontario, Lake Michigan and Lake Störvindeln were shown and the  $t_{1/2}$  values of T-DDT were calculated. The  $t_{1/2}$  values were 9, 11, 8 and 7 years, respectively, in Lake Biwa, Lake Ontario, Lake Michigan and Lake Störvindeln. There were no wide differences in the  $t_{1/2}$  values between the four lakes.
3. Composition of T-DDT in fish from lakes and rivers in the world was compared for the 25 lakes in 15 countries of Europe, America, Asia and Africa from 1996 to 2008 and for the 16 rivers in 8 countries of Europe, America, Asia, Africa and Oceania from 2000 to 2009. Similarly, composition of T-HCH in fish from lakes and rivers in the world was compared for the 16 lakes in 8 countries of Europe, America, Asia and Africa from 1996 to 2008 and for the 11 rivers in 5 countries of Europe and Asia from 2001 to 2006.

## 2. Residue of T-DDT and T-HCH in fish from lakes and rivers in the world

Residue of T-DDT and T-HCH in fish from lakes and rivers in the world (Survey years: 1995~2009) was reviewed from literatures in the past. The residue data were summarized in Table 1 for the lakes [22-31, 35-70] and in Table 2 for the rivers [40, 51, 68, 71-101].

No.	Species	n <sup>a</sup>	Analyte	Lake	Year	Country	T-HCH <sup>b</sup>	T-DDT <sup>c</sup>	References
L-1	Japanese dace	5	Muscle	Lake Biwa	2000	Japan	3 ng/g wet wt	13 ng/g wet wt	Ministry of the Environment, Japan (2002)
L-2	Japanese dace	5	Muscle	Lake Biwa	2001	Japan	2 ng/g wet wt	10 ng/g wet wt	Ministry of the Environment, Japan (2003)
L-3	Japanese dace	5	Muscle	Lake Biwa	2002	Japan	1.79 ng/g wet wt	6.5 ng/g wet wt	Ministry of the Environment, Japan (2004)
L-4	Japanese dace	5	Muscle	Lake Biwa	2003	Japan	0.97 ng/g wet wt	8.0 ng/g wet wt	Ministry of the Environment, Japan (2005)
L-5	Japanese dace	5	Muscle	Lake Biwa	2004	Japan	0.55 ng/g wet wt	8.4 ng/g wet wt	Ministry of the Environment, Japan (2006)
L-6	Japanese dace	5	Muscle	Lake Biwa	2005	Japan	0.29 ng/g wet wt	9.3 ng/g wet wt	Ministry of the Environment, Japan (2007)

No.	Species	n <sup>a</sup>	Analyte	Lake	Year	Country	T-HCH <sup>b</sup>	T-DDT <sup>c</sup>	References
L-7	Japanese dace	5	Muscle	Lake Biwa	2006	Japan	0.90 ng/g wet wt	8.3 ng/g wet wt	Ministry of the Environment, Japan (2008)
L-8	Raibow trout (male)	4	Muscle	Lake Mashu	2003	Japan	2.98 ng/g wet wt	1.49 ng/g wet wt	Takazawa et al. (2005)
L-9	Raibow trout (female)	6	Muscle	Lake Mashu	2003	Japan	2.71 ng/g wet wt	1.72 ng/g wet wt	Takazawa et al. (2005)
L-10	Japanese dace (male)	3	Muscle	Lake Mashu	2003	Japan	1.75 ng/g wet wt	0.66 ng/g wet wt	Takazawa et al. (2005)
L-11	Japanese dace (female)	6	Muscle	Lake Mashu	2003	Japan	2.45 ng/g wet wt	0.56 ng/g wet wt	Takazawa et al. (2005)
L-12	Raibow trout (male)	3	Muscle	Lake Mashu	2002	Japan	3.23 ng/g wet wt	3.50 ng/g wet wt	Takazawa et al. (2005)
L-13	Raibow trout (female)	9	Muscle	Lake Mashu	2002	Japan	2.19 ng/g wet wt	1.27 ng/g wet wt	Takazawa et al. (2005)
L-14	Unkown	6	Whole	Taihu Lake Region	1999~2000	China	46 ng/g wet wt	12 ng/g wet wt	Feng et al. (2003)
L-15	Carp	3	Whole	Lake Tai	2000	China	64 ng/g fat wt	980 ng/g fat wt	Nakata et al. (2005)
L-16	Topmouth culter	3	Whole	Lake Tai	2000	China	67 ng/g fat wt	750 ng/g fat wt	Nakata et al. (2005)
L-17	Spotted steed	3	Whole	Lake Tai	2000	China	75 ng/g fat wt	700 ng/g fat wt	Nakata et al. (2005)
L-18	Catfish	3	Whole	Lake Tai	2000	China	68 ng/g fat wt	1000 ng/g fat wt	Nakata et al. (2005)
L-19	<i>Gymnocypris namensis</i>	4	Muscle	Nam Co Lake	2005	China	2.57 ng/g wet wt	17.2 ng/g wet wt	Yang et al. (2007)
L-20	<i>Gymnocypris waddellii</i>	4	Muscle	Yamdro Lake	2005	China	1.56 ng/g wet wt	2.76 ng/g wet wt	Yang et al. (2007)
L-21	<i>C. auratus</i>	8	Edible part	Gaobeidian Lake (Beijing)	2006	China	6.41 ng/g wet wt	21.96 ng/g wet wt	Li et al. (2008)
L-22	<i>M. anguillicaudatus</i>	5	Edible part	Gaobeidian Lake (Beijing)	2006	China	2.61 ng/g wet wt	14.08 ng/g wet wt	Li et al. (2008)
L-23	<i>H. leuciscultures</i>	8	Edible part	Gaobeidian Lake (Beijing)	2006	China	11.14 ng/g wet wt	84.4 ng/g wet wt	Li et al. (2008)
L-24	Herbivorous	9	Muscle	Songkhla Lake (Thale Luang)	1997	Thailand		170 ng/g fat wt	Kumblad et al. (2001)
L-25	Herbivorous	10	Muscle	Songkhla Lake (Thale Sap)	1997	Thailand		36 ng/g fat wt	Kumblad et al. (2001)
L-26	Herbivorous	8	Muscle	Songkhla Lake (Thale Sap Songkhla)	1997	Thailand		35 ng/g fat wt	Kumblad et al. (2001)
L-27	<i>Channa striata</i>	64	Muscle	Kolleru Lake	Unkown	India			Amaraneri & Pillala (2001)
L-28	<i>Channa striata</i>	56	Liver	Kolleru Lake	Unkown	India			Amaraneri & Pillala (2001)
L-29	<i>Catla catla</i>	58	Muscle	Kolleru Lake	Unkown	India			Amaraneri & Pillala (2001)
L-30	<i>Catla catla</i>	38	Liver	Kolleru Lake	Unkown	India			Amaraneri & Pillala (2001)
L-31	<i>P. phuturio</i>	2	Whole	Haleji Lake	1999	Pakistan		4.55 ng/g wet wt	Sanpera et al. (2002)
L-32	<i>C. lalia</i>	3	Whole	Haleji Lake	1999	Pakistan		5.58 ng/g wet wt	Sanpera et al. (2002)
L-33	<i>G. giuris</i>	1	Whole	Haleji Lake	1999	Pakistan		5.94 ng/g wet wt	Sanpera et al. (2002)

## 20 Pesticides – Recent Trends in Pesticide Residue Assay

No.	Species	n <sup>a</sup>	Analyte	Lake	Year	Country	T-HCH <sup>b</sup>	T-DDT <sup>c</sup>	References
L-34	Several kinds	81	Edible part	Lake Jarun	2000	Croatia	0.40 ng/g wet wt*	0.80 ng/g wet wt*	Bosnir et al. (2007)
L-35	Arctic char	25	Muscle	Lake Ellasjøen	1996	Norway		60 ng/g wet wt	Evenset et al. (2004)
L-36	Arctic char	12	Muscle	Lake Øyangen	1996	Norway		4.3 ng/g wet wt	Evenset et al. (2004)
L-37	Whitefish (Female) 100-200g	13	Muscle	Lake Stuorajavri	2005	Norway	ND	0.35 ng/g wet wt	Christensen et al. (2007)
L-38	Whitefish (Male) 100-200g	10	Muscle	Lake Stuorajavri	2005	Norway	ND	0.41 ng/g wet wt	Christensen et al. (2007)
L-39	Pike	5	Muscle	Lake Stuorajavri	2005	Norway	ND	0.24 ng/g wet wt	Christensen et al. (2007)
L-40	Brown trout	1	Muscle	L. Tuma (remote alpine lake)	2003	Switzerland		1100 ng/g fat wt (+op')	Schmid et al. (2007)
L-41	Brown trout	1	Muscle	L. Moesola (remote alpine lake)	2003	Switzerland		680 ng/g fat wt(+op')	Schmid et al. (2007)
L-42	Lake trout	1	Muscle	L. Diavolezza (remote alpine lake)	2003	Switzerland		130 ng/g fat wt(+op')	Schmid et al. (2007)
L-43	Catfish	8	Muscle	Lake Trasimeno	1998	Italy	14.3 ng/g fat wt	216 ng/g fat wt	Elia et al. (2006)
L-44	Mullet ( <i>Lisa aurata</i> )	13	Muscle	Lake Ganzirri	2001	Italy		3.8 ng/g wet wt (Max.)	Licata et al. (2003)
L-45	Landlocked	5	Whole	Lake Maggiore	2002-2004	Italy		2500 ng/g fat wt	Bettinetti et al. (2006)
L-46	Whitefish	5	Whole	Lake Maggiore	2002-2004	Italy		1370 ng/g fat wt	Bettinetti et al. (2006)
L-47	Perch	5	Whole	Lake Maggiore	2002-2004	Italy		1860 ng/g fat wt	Bettinetti et al. (2006)
L-48	Chub	5	Whole	Lake Maggiore	2002-2004	Italy		1190 ng/g fat wt	Bettinetti et al. (2006)
L-49	Rudd	5	Whole	Lake Maggiore	2002-2004	Italy		2770 ng/g fat wt	Bettinetti et al. (2006)
L-50	Tench	5	Whole	Lake Maggiore	2002-2004	Italy		2720 ng/g fat wt	Bettinetti et al. (2006)
L-51	Perch	1	Muscle	Bolsena Lake	2002	Italy	0.02 ng/g wet wt	2.26 ng/g wet wt	Orban et al. (2007)
L-52	Perch	1	Muscle	Bracciano Lake	2002	Italy	0.09 ng/g wet wt	0.38 ng/g wet wt	Orban et al. (2007)
L-53	Perch	1	Muscle	Salto Lake	2002	Italy	0.03 ng/g wet wt	0.61 ng/g wet wt	Orban et al. (2007)
L-54	Pelagic landlocked shad	1	Muscle	Lake Como (Como branch)	2006	Italy		1010 ng/g fat wt	Bettinetti et al. (2008)
L-55	Pelagic landlocked shad	1	Muscle	Lake Como (Como branch)	2007	Italy		840 ng/g fat wt	Bettinetti et al. (2008)
L-56	Pelagic landlocked shad	1	Muscle	Lake Como (Lecco branch)	2007	Italy		610 ng/g fat wt	Bettinetti et al. (2008)
L-57	Pelagic landlocked shad	1	Muscle	Lake Iseo	2007	Italy		570 ng/g fat wt	Bettinetti et al. (2008)

No.	Species	n <sup>a</sup>	Analyte	Lake	Year	Country	T-HCH <sup>b</sup>	T-DDT <sup>c</sup>	References
L-58	Pikeperch	4	Muscle	Beysehir Lake	Unkown	Turkey	70 ng/g wet wt	27 ng/g wet wt	Aktumsek et al. (2002)
L-59	Carp	17	Muscle	Sir Dam Lake	2003	Turkey	0.21 ng/g wet wt*	14.4 ng/g wet wt*	Erdogrul et al. (2005)
L-60	<i>Oreochromis niloticus</i>	4	Muscle	Lake Burullus	2006	Egypt	1.88 ng/g wet wt	5.13 ng/g wet wt	Said et al. (2008)
L-61	<i>Clarias sp.</i>	4	Muscle	Lake Burullus	2006	Egypt	9.83 ng/g wet wt	12.54 ng/g wet wt	Said et al. (2008)
L-62	<i>Bagrus meridionalis</i>	4	Muscle	Lake Malawi	1996, 1997	East Africa		13.1 ng/g wet wt	Kidd et al. (2001)
L-63	<i>Buccochromis nototaenia</i>	2	Muscle	Lake Malawi	1996, 1997	East Africa		3.4 ng/g wet wt	Kidd et al. (2001)
L-64	<i>Clarius sp.</i>	1	Muscle	Lake Malawi	1996, 1997	East Africa		1.4 ng/g wet wt	Kidd et al. (2001)
L-65	<i>Engraulicyprus sardella</i>	6	Whole	Lake Malawi	1996, 1997	East Africa		4.5 ng/g wet wt	Kidd et al. (2001)
L-66	<i>Genyochromis mento</i>	5	Whole	Lake Malawi	1996, 1997	East Africa		1.0 ng/g wet wt	Kidd et al. (2001)
L-67	<i>Labeotropheus fuelleborni</i>	6	Whole	Lake Malawi	1996, 1997	East Africa		1.1 ng/g wet wt	Kidd et al. (2001)
L-68	<i>Boulengerochromis microlepis</i>	1	Whole	Lake Tanganyika (North end)	1999	Burundi	288.2 ng/g fat wt	794.7 ng/g fat wt	Manirakiza et al. (2002)
L-69	<i>Chrysichthys sianenna</i>	1	Whole	Lake Tanganyika (North end)	1999	Burundi	90.6 ng/g fat wt	339.3 ng/g fat wt	Manirakiza et al. (2002)
L-70	<i>Oreochromis niloticus</i>	1	Whole	Lake Tanganyika (North end)	1999	Burundi	66.2 ng/g fat wt	393.1 ng/g fat wt	Manirakiza et al. (2002)
L-71	<i>Limnothrissa miodon</i>	1	Whole	Lake Tanganyika (North end)	1999	Burundi	21.2 ng/g fat wt	60.7 ng/g fat wt	Manirakiza et al. (2002)
L-72	<i>Stolothrissa tanganyikae</i>	1	Whole	Lake Tanganyika (North end)	1999	Burundi	55.1 ng/g fat wt	95.7 ng/g fat wt	Manirakiza et al. (2002)
L-73	Nile tilapia	43	Edible part	Lake Victoria (Napoleon Gulf)	1998	Uganda		1.39 ng/g wet wt	Kasozi et al. (2006)
L-74	Nile perch	37	Edible part	Lake Victoria (Napoleon Gulf)	1998	Uganda		1.67 ng/g wet wt	Kasozi et al. (2006)
L-75	Nile tilapia and Nile perch		Muscle	Lake Victoria (Kome Island)	1999	Tanzania		20 ng/g wet wt	Henry & Kishimba (2006)
L-76	Nile tilapia and Nile perch		Muscle	Lake Victoria (Katunguru)	1999	Tanzania		15 ng/g wet wt	Henry & Kishimba (2006)

## 22 Pesticides – Recent Trends in Pesticide Residue Assay

No.	Species	n <sup>a</sup>	Analyte	Lake	Year	Country	T-HCH <sup>b</sup>	T-DDT <sup>c</sup>	References
L-77	Nile tilapia	10	Muscle	Lake Taabo	Unkown	Cote d'Ivoire	225.8 ng/g dry wt	124.1 ng/g dry wt	Roche et al. (2007)
L-78	African catfish	7	Muscle	Lake Taabo	Unkown	Cote d'Ivoire	130.0 ng/g dry wt	94.6 ng/g dry wt	Roche et al. (2007)
L-79	Raibow trout	3	Muscle	Rae Lake (Sierra Nevada)	1995, 1996	USA		1.24 ng/g wet wt	Ohyama et al. (2004)
L-80	Raibow trout	6	Muscle	Blue Lake (Sierra Nevada)	1995, 1996	USA		2.13 ng/g wet wt	Ohyama et al. (2004)
L-81	Raibow trout	5	Muscle	Pear Lake (Sierra Nevada)	1995, 1996	USA		9.99 ng/g wet wt	Ohyama et al. (2004)
L-82	Raibow trout	2	Muscle	Donner Lake (Sierra Nevada)	1995, 1996	USA		3.71 ng/g wet wt	Ohyama et al. (2004)
L-83	Raibow trout	4	Muscle	Huntington Lake (Sierra Nevada)	1995, 1996	USA		30.55 ng/g wet wt	Ohyama et al. (2004)
L-84	Raibow trout	5	Muscle	Lake Berryessa (Sierra Nevada)	1995, 1996	USA		13.01 ng/g wet wt	Ohyama et al. (2004)
L-85	Lake trout	10	Muscle	Lake Chelan (Wapato Basin)	2003	USA		943 ng/g wet wt	Washington State Department of Ecology (2005)
L-86	Burbot	7	Muscle	Lake Chelan (Wapato Basin)	2003	USA		315 ng/g wet wt	
L-87	Kokanee	7	Muscle	Lake Chelan (Wapato Basin)	2003	USA		57 ng/g wet wt	
L-88	Raibow trout	3	Muscle	Lake Chelan (Wapato Basin)	2003	USA		14 ng/g wet wt	
L-89	Burbot	3	Muscle	Lake Chelan (Lucerne Basin)	2003	USA		22 ng/g wet wt	
L-90	Raibow trout	1	Muscle	Roses Lake	2003	USA		96 ng/g wet wt	Washington State Department of Ecology (2007)
L-91	Black Crappie	1	Muscle	Roses Lake	2003	USA		32 ng/g wet wt	
L-92	Lg Scale Sucker	15	Muscle	Vancouver Lake	2005-2006	USA	ND	23 ng/g wet wt	
L-93	Common Carp	15	Muscle	Vancouver Lake	2006	USA	ND	65 ng/g wet wt	
L-94	Lg Mouth Bass	5	Muscle	Vancouver Lake	2006	USA	ND	42 ng/g wet wt	McIntyre & Beauchamp (2007)
L-95	N. pikeminnow (Large size)	10	Whole	Lake Washington	2001-2003	USA		258 ng/g wet wt	



No.	Species	n <sup>a</sup>	Analyte	Lake	Year	Country	T-HCH <sup>b</sup>	T-DDT <sup>c</sup>	References
L-96	Cutthroat trout (Large size)	10	Whole	Lake Washington	2001-2003	USA		168 ng/g wet wt	McIntyre & Beauchamp (2007)
L-97	Yellow perch (Large size)	9	Whole	Lake Washington	2001-2003	USA		59 ng/g wet wt	McIntyre & Beauchamp (2007)
L-98	Smallmouth bass (Large size)	3	Whole	Lake Washington	2001-2003	USA		63 ng/g wet wt	McIntyre & Beauchamp (2007)
L-99	Lake trout	10	Muscle	Kusawa Lake	1993	Canada	1.21 ng/g wet wt	40.85 ng/g wet wt	Ryan et al. (2005)
L-100	Lake trout	14	Muscle	Kusawa Lake	1999	Canada	1.68 ng/g wet wt	122.43 ng/g wet wt	Ryan et al. (2005)
L-101	Lake trout	9	Muscle	Kusawa Lake	2001	Canada	0.91 ng/g wet wt	49.71 ng/g wet wt	Ryan et al. (2005)
L-102	Lake trout	10	Muscle	Kusawa Lake	2002	Canada	0.62 ng/g wet wt	23.51 ng/g wet wt	Ryan et al. (2005)
L-103	Lake trout	24	Muscle	Lake Laberge	1993	Canada	4.69 ng/g wet wt	360.87 ng/g wet wt	Ryan et al. (2005)
L-104	Lake trout	13	Muscle	Lake Laberge	1996	Canada	6.50 ng/g wet wt	205.54 ng/g wet wt	Ryan et al. (2005)
L-105	Lake trout	5	Muscle	Lake Laberge	2000	Canada	2.30 ng/g wet wt	82.96 ng/g wet wt	Ryan et al. (2005)
L-106	Lake trout	16	Muscle	Lake Laberge	2001	Canada	0.80 ng/g wet wt	75.09 ng/g wet wt	Ryan et al. (2005)
L-107	Lake trout	5	Muscle	Lake Laberge	2002	Canada	1.58 ng/g wet wt	43.56 ng/g wet wt	Ryan et al. (2005)
L-108	Lake trout	8	Muscle	Lake Laberge	2003	Canada	0.54 ng/g wet wt	55.81 ng/g wet wt	Ryan et al. (2005)
L-109	Trahira ( <i>Hoplias malabaricus</i> )	10	Muscle	Ponta Grossa Lake	2005	Brazil		92.3 ng/g dry wt	Miranda et al. (2008)
L-110	Trahira ( <i>Hoplias malabaricus</i> )	10	Liver	Ponta Grossa Lake	2005	Brazil		54.68 ng/g dry wt	Miranda et al. (2008)
L-111	Japanese dace	5	Muscle	Lake Biwa	2007	Japan	0.51 ng/g wet wt	6.9 ng/g wet wt	Ministry of the Environment, Japan (2009)
L-112	Japanese dace	5	Muscle	Lake Biwa	2008	Japan	0.68 ng/g wet wt	8.0 ng/g wet wt	Ministry of the Environment, Japan (2010)
L-113	Japanese dace	5	Muscle	Lake Biwa	2009	Japan	0.41 ng/g wet wt	8.0 ng/g wet wt	Ministry of the Environment, Japan (2011)
L-114	Semutundu ( <i>Bagrus docmac</i> )		Muscle	Lake Edward	Unkown	Uganda		33 ng/g wet wt	Ssebugere et al. (2009)
L-115	Mamba ( <i>Protopterus aethiopus</i> )		Muscle	Lake Edward	Unkown	Uganda		29 ng/g wet wt	Ssebugere et al. (2009)
L-116	Enjunguri ( <i>Haplochromis nigripinnis</i> )		Muscle	Lake Edward	Unkown	Uganda		ND	Ssebugere et al. (2009)
L-117	Nile tilapia ( <i>Oreochromis niloticus</i> )		Muscle	Lake Edward	Unkown	Uganda		33 ng/g wet wt	Ssebugere et al. (2009)



No.	Species	n <sup>a</sup>	Analyte	Lake	Year	Country	T-HCH <sup>b</sup>	T-DDT <sup>c</sup>	References
L-118	Male ( <i>Clarias gariepinus</i> )		Muscle	Lake Edward	Unkown	Uganda		ND	Ssebugere et al. (2009)
L-119	Tilapia and Catfish	13	Edible part	Lake Volta, Lake Bosumtwi, Weija Lake	2008	Ghana	0.72 ng/g fat wt	329.4 ng/g fat wt	Adu-Kumi et al. (2010)
L-120	6 kinds of fish	60	Muscle	Lakes (n=8) in Tibetan Plateau	2006-2007	China	0.55 ng/g wet wt	4.0 ng/g wet wt (+op')	Yang et al. (2010)
L-121	Common carp	23	Unknown	Baiyangdian Lake	2008	China	0.38 ng/g wet wt	1.28 ng/g wet wt	Dai et al. (2011)
L-122	Crucian carp	25	Unknown	Baiyangdian Lake	2008	China	0.47 ng/g wet wt	1.03 ng/g wet wt	Dai et al. (2011)
L-123	Crucian carp	1	Muscle	Lake Como	2007	Italy	1.0 ng/g dry wt	1.03 ng/g dry wt (+op')	Villa et al. (2011)
L-124	White fish	1	Muscle	Lake Como	2007	Italy	7.35 ng/g dry wt	12.4 ng/g dry wt (+op')	Villa et al. (2011)
L-125	Pike	1	Muscle	Lake Como	2007	Italy	0.4 ng/g dry wt	4.89 ng/g dry wt (+op')	Villa et al. (2011)
L-126	Chub	1	Muscle	Lake Como	2007	Italy	ND	5.89 ng/g dry wt (+op')	Villa et al. (2011)
L-127	Perch	1	Muscle	Lake Como	2007	Italy	0.7 ng/g dry wt	7.75 ng/g dry wt (+op')	Villa et al. (2011)
L-128	Pikeperch	1	Muscle	Lake Como	2007	Italy	0.57 ng/g dry wt	10.4 ng/g dry wt (+op')	Villa et al. (2011)

<sup>a</sup> No. of analyzed samples; Mean (n≥2) or single determination values (n=1) are listed for PCB, T-HCH and T-DDT ( \* Median value)

<sup>b</sup> T-HCH =  $\alpha$ -HCH +  $\beta$ -HCH +  $\gamma$ -HCH

<sup>c</sup> T-DDT = pp'-DDE + pp'-DDD + PP'-DDT

**Table 1.** Concentrations of T-HCH and T-DDT in fish from lakes in the world

No.	Species	n <sup>a</sup>	Analyte	River	Year	Country	T-HCH <sup>b</sup>	T-DDT <sup>c</sup>	References
R-1	Color gudgeon	10	Whole	Guanting Reservoir	2002	China	7.15 ng/g wet wt	9.23 ng/g wet wt	Sun et al. (2005)
R-2	Feral carp	10	Whole	Guanting Reservoir	2002	China	0.72 ng/g wet wt	5.04 ng/g wet wt	Sun et al. (2005)
R-3	White fish	1	Unknown	Qiantang River (Downstream)	2005	China	3.96 ng/g wet wt	13.51 ng/g wet wt	Zhou et al. (2007)
R-4	Crucian carp	1	Unknown	Qiantang River (Downstream)	2005	China	3.84 ng/g wet wt	5.64 ng/g wet wt	Zhou et al. (2007)
R-5	Perch	1	Unknown	Qiantang River (Downstream)	2005	China	2.62 ng/g wet wt	8.34 ng/g wet wt	Zhou et al. (2007)
R-6	Snake head mullet	1	Unknown	Qiantang River (Upstream)	2005	China	3.18 ng/g wet wt	5.01 ng/g wet wt	Zhou et al. (2007)
R-7	Bulltrout	1	Unknown	Qiantang River (Upstream)	2005	China	2.85 ng/g wet wt	2.30 ng/g wet wt	Zhou et al. (2007)
R-8	<i>Ptychobarbus dipogon</i>	3	Muscle	Lhasa River	2005	China	0.286 ng/g wet wt	2.07 ng/g wet wt	Yang et al. (2007)
R-9	<i>Schizopygopsis younhusbandi</i>	3	Muscle	Lhasa River	2005	China	0.75 ng/g wet wt	2.99 ng/g wet wt	Yang et al. (2007)
R-10	<i>C. auratus</i>	5	Edible part	Huairou Reservoir (Beijing)	2006	China	0.34 ng/g wet wt	7.53 ng/g wet wt	Li et al. (2008)
R-11	<i>M. anguillicaudatus</i>	6	Edible part	Huairou Reservoir (Beijing)	2006	China	5.42 ng/g wet wt	44.17 ng/g wet wt	Li et al. (2008)

No.	Species	n <sup>a</sup>	Analyte	River	Year	Country	T-HCH <sup>b</sup>	T-DDT <sup>c</sup>	References
R-12	<i>P. fulvidraco</i>	4	Edible part	Huairou Reservoir (Beijing)	2006	China	1.93 ng/g wet wt	34.5 ng/g wet wt	Li et al. (2008)
R-13	Crucian carp	1	Edible part	Qiantang River (Jinhua)	2006	China	253 ng/g fat wt	514 ng/g fat wt	Zhou et al. (2008)
R-14	White fish	1	Edible part	Qiantang River (Jinhua)	2006	China	222 ng/g fat wt	315 ng/g fat wt	Zhou et al. (2008)
R-15	Crucian carp	1	Edible part	Qiantang River (Fuchunjiang)	2006	China	1054 ng/g fat wt	1408 ng/g fat wt	Zhou et al. (2008)
R-16	White fish	1	Edible part	Qiantang River (Fuchunjiang)	2006	China	152 ng/g fat wt	1330 ng/g fat wt	Zhou et al. (2008)
R-17	Crucian carp	1	Edible part	Qiantang River (Fuyang)	2006	China	577 ng/g fat wt	155 ng/g fat wt	Zhou et al. (2008)
R-18	White fish	1	Edible part	Qiantang River (Fuyang)	2006	China	467 ng/g fat wt	715 ng/g fat wt	Zhou et al. (2008)
R-19	Crucian carp	1	Edible part	Qiantang River (Hangzhou)	2006	China	936 ng/g fat wt	1199 ng/g fat wt	Zhou et al. (2008)
R-20	White fish	1	Edible part	Qiantang River (Hangzhou)	2006	China	844 ng/g fat wt	3008 ng/g fat wt	Zhou et al. (2008)
R-21	Many kinds of fish	19	Whole	Pearl River Estuary	2004	China	0.20 ng/g wet wt*	77 ng/g wet wt*	Guo et al. (2008)
R-22	Kissing gourami	2	Whole	Ciliwung River	2003	Indonesia	24 ng/g fat wt	800 ng/g fat wt	Sudaryanto et al. (2007)
R-23	Common carp	1	Whole	Ciliwung River	2003	Indonesia	3.4 ng/g fat wt	37 ng/g fat wt	Sudaryanto et al. (2007)
R-24	Tilapia mossambique	1	Whole	Ciliwung River	2003	Indonesia	6.0 ng/g fat wt	1100 ng/g fat wt	Sudaryanto et al. (2007)
R-25	Walking catfish	4	Whole	Ciliwung River	2003	Indonesia	11 ng/g fat wt	610 ng/g fat wt	Sudaryanto et al. (2007)
R-26	Unknown (n=10)		Unknown	Rivers and streams in Kumaun Himalayas	1999	India	1 ng/g wet wt	13 ng/g wet wt	Sarkar et al. (2003)
R-27	<i>Scatophagus argus</i>	3	Muscle	Ferok River	2003	India	0.72 ng/g wet wt	3.11 ng/g wet wt	Sarkar et al. (2006)
R-28	<i>Platicephalus sp.</i>	3	Muscle	Korappuzha River	2003	India	5.4 ng/g wet wt	ND	Sarkar et al. (2006)
R-29	<i>Etroplus suratensis</i>	3	Muscle	Pukatri River	2003	India	0.06 ng/g wet wt	0.44 ng/g wet wt	Sarkar et al. (2006)
R-30	Burbot	3	Liver	Pechora River	1988	Russia	11 ng/g wet wt	57 ng/g wet wt	Zhulidov et al. (2002)
R-31	Burbot	3	Liver	Pechora River	1994	Russia	4 ng/g wet wt	9 ng/g wet wt	Zhulidov et al. (2002)
R-32	Several kinds	103	Edible part	Sava River	2000	Croatia	0.70 ng/g wet wt*	1.80 ng/g wet wt*	Bosnir et al. (2007)
R-33	4 kinds of fish	4	Muscle	Dniester River (Upstream)	2001	Moldova		17.3 ng/g wet wt	Sapozhnikova et al. (2005)
R-34	5 kinds of fish	5	Muscle	Dniester River (Downstream)	2001	Moldova		10.2 ng/g wet wt	Sapozhnikova et al. (2005)
R-35	European river lamprey	31	Whole	Rivers flowing (n=8) to Bothnian Bay	Unkown	Finland	3.03 ng/g wet wt	46 ng/g wet wt	Merivirta et al. (2006)
R-36	European river lamprey	19	Whole	Rivers (n=5) flowing to Bothnian Sea	Unkown	Finland	4.20 ng/g wet wt	68 ng/g wet wt	Merivirta et al. (2006)
R-37	Chub	3	Muscle	River Nestos (Paranesti)	2004	Greece	0.27 ng/g wet wt	0.40 ng/g wet wt	Christoforidis et al. (2008)

## 26 Pesticides – Recent Trends in Pesticide Residue Assay

No.	Species	n <sup>a</sup>	Analyte	River	Year	Country	T-HCH <sup>b</sup>	T-DDT <sup>c</sup>	References
R-38	Chub	3	Muscle	River Nestos (Kommnina)	2004	Greece	0.10 ng/g wet wt	ND	Christoforidis et al. (2008)
R-39	Barbel	3	Muscle	River Nestos (Paranesti)	2004	Greece	0.91 ng/g wet wt	0.47 ng/g wet wt	Christoforidis et al. (2008)
R-40	Barbel	3	Muscle	River Nestos (Kommnina)	2004	Greece	0.15 ng/g wet wt	0.25 ng/g wet wt	Christoforidis et al. (2008)
R-41	Common trout	5	Muscle	River Turia	2000	Spain		4.3 ng/g wet wt	Bordajandi et al. (2003)
R-42	European eel	11	Muscle	River Turia	2000	Spain		45.3 ng/g wet wt	Bordajandi et al. (2003)
R-43	Brown trout	28	Whole	Two rivers in Cantabria	2001	Spain	0.55 ng/g dry wt	20.2 ng/g dry wt	Guitart et al. (2005)
R-44	Eurasian minnow	17	Whole	Two rivers in Cantabria	2001	Spain	1.04 ng/g dry wt	23.0 ng/g dry wt	Guitart et al. (2005)
R-45	European eel	16	Whole	Two rivers in Cantabria	2001	Spain	0.66 ng/g dry wt	39.4 ng/g dry wt	Guitart et al. (2005)
R-46	Barbel	3	Whole	Ebro River Basin (Presa de Pina)	2003	Spain		35.9 ng/g dry wt	Lacorte et al. (2006)
R-47	Bleak	6	Whole	Ebro River Basin (Presa de Pina)	2003	Spain		71.9 ng/g dry wt	Lacorte et al. (2006)
R-48	Common carp	1	Whole	Ebro River Basin (Flix)	2003	Spain		983 ng/g dry wt	Lacorte et al. (2006)
R-49	Bleak	3	Whole	Ebro River Basin (Flix)	2003	Spain		487 ng/g dry wt	Lacorte et al. (2006)
R-50	Barbel	2	Muscle	Cinca River (Upstream)	2002	Spain		31 ng/g wet wt (+op')	De la Cal et al. (2008)
R-51	Barbel	2	Muscle	Cinca River (Downstream)	2002	Spain		780 ng/g wet wt (+op')	De la Cal et al. (2008)
R-52	Bleak	1	Whole	Cinca River (Upstream)	2002	Spain		5 ng/g wet wt (+op')	De la Cal et al. (2008)
R-53	Bleak	2	Whole	Cinca River (Downstream)	2002	Spain		508 ng/g wet wt (+op')	De la Cal et al. (2008)
R-54	Tilapia zilli	2	Unknown	Ogba River	Unkown	Nigeria		56 ng/g wet wt	Ize-Iyamu et al. (2007)
R-55	Catfish	2	Unknown	Ogba River	Unkown	Nigeria		106 ng/g wet wt	Ize-Iyamu et al. (2007)
R-56	Tilapia zilli	2	Unknown	Ovia River	Unkown	Nigeria		61 ng/g wet wt	Ize-Iyamu et al. (2007)
R-57	Catfish	2	Unknown	Ovia River	Unkown	Nigeria		115 ng/g wet wt	Ize-Iyamu et al. (2007)
R-58	Tilapia zilli	2	Unknown	Ikoru Riber	Unkown	Nigeria		20 ng/g wet wt	Ize-Iyamu et al. (2007)
R-59	Catfish	2	Unknown	Ikoru Riber	Unkown	Nigeria		34 ng/g wet wt	Ize-Iyamu et al. (2007)
R-60	Smallmouth bass	3	Whole	Willamette River (Lower Superfund)	2000	USA (Oregon)	< 8 ng/g wet wt	320 ng/g wet wt	Sethajintanin et al. (2004)
R-61	Common carp	3	Whole	Willamette River (Lower Superfund)	2000	USA (Oregon)	< 8 ng/g wet wt	97 ng/g wet wt	Sethajintanin et al. (2004)
R-62	Carp	3	Muscle	Okanogan River (Oroville)	2001	USA		336 ng/g wet wt	Washington State Department of Ecology (2003)
R-63	Mountain whitefish	3	Muscle	Okanogan River (Oroville)	2001	USA		350 ng/g wet wt	
R-64	Smallmouth bass	3	Muscle	Okanogan River (Oroville)	2001	USA		157 ng/g wet wt	

No.	Species	n <sup>a</sup>	Analyte	River	Year	Country	T-HCH <sup>b</sup>	T-DDT <sup>c</sup>	References
R-65	Akupa sleeper fish	1	Unknown	Hanalei River	2001	USA (Hawaii)	< 1 ng/g wet wt	< 2 ng/g wet wt	Orazio et al. (2007)
R-66	Chinook salmon		whole	Lower Columbia Estuary	2001-2002	USA		1800-27000 ng/g fat wt (+op')	Johnson et al. (2007)
R-67	Largemouth bass	10	whole	Mobile River basin (Lavaca)	2004	USA (Alabama)		24.9 ng/g wet wt	Hinck et al. (2009)
R-68	Largemouth bass	10	whole	Mobile River basin (Mcintosh)	2004	USA (Alabama)		6946 ng/g wet wt	Hinck et al. (2009)
R-69	Largemouth bass	10	whole	Mobile River basin (Bucks)	2004	USA (Alabama)		92.8 ng/g wet wt	Hinck et al. (2009)
R-70	Largemouth bass	8	whole	Mobile River basin	2004	USA		53.84 ng/g wet wt	Hinck et al. (2008)
R-71	Largemouth bass	6	whole	Apalachicola-Chattahoochee-Flint River Basin	2004	USA		87.45 ng/g wet wt	Hinck et al. (2008)
R-72	Largemouth bass	6	whole	Savannah River Basin	2004	USA		18.98 ng/g wet wt	Hinck et al. (2008)
R-73	Largemouth bass	6	whole	Pee Dee River Basin	2004	USA		37.84 ng/g wet wt	Hinck et al. (2008)
R-74	Carp	8	whole	Mobile River basin	2004	USA		41.58 ng/g wet wt	Hinck et al. (2008)
R-75	Carp	6	whole	Apalachicola-Chattahoochee-Flint River Basin	2004	USA		90.64 ng/g wet wt	Hinck et al. (2008)
R-76	Carp	6	whole	Savannah River Basin	2004	USA		16.42 ng/g wet wt	Hinck et al. (2008)
R-77	Carp	6	whole	Pee Dee River Basin	2004	USA		20.42 ng/g wet wt	Hinck et al. (2008)
R-78	Chub	10	Muscle	River Elbe (Downstream Pardubice)	2004	Czech Republic	24 ng/g fat wt	2850 ng/g fat wt (+op')	Randak et al. (2009)
R-79	Chub	8	Muscle	River Elbe (Downstream Neratovice)	2004	Czech Republic	486 ng/g fat wt	4830 ng/g fat wt (+op')	Randak et al. (2009)
R-80	Chub	4	Muscle	River Elbe (Downstream Usti nad Labem)	2004	Czech Republic	53 ng/g fat wt	6480 ng/g fat wt (+op')	Randak et al. (2009)
R-81	Chub ( <i>Leuciscus cephalus</i> )	10	Muscle	Svratka River (Modřice)	Apr.-2007	Czech Republic	1.0 ng/g wet wt	34.9 ng/g wet wt (+op')	Lána et al. (2010)
R-82	Chub ( <i>Leuciscus cephalus</i> )	10	Muscle	Svratka River (Modřice)	Oct.-2007	Czech Republic	0.7 ng/g wet wt	29.4 ng/g wet wt (+op')	Lána et al. (2010)
R-83	Chub ( <i>Leuciscus cephalus</i> )	9	Muscle	Svratka River (Rajhradice)	Apr.-2007	Czech Republic	0.9 ng/g wet wt	40.0 ng/g wet wt (+op')	Lána et al. (2010)
R-84	Chub ( <i>Leuciscus cephalus</i> )	11	Muscle	Svratka River (Rajhradice)	Oct.-2007	Czech Republic	2.6 ng/g wet wt	28.8 ng/g wet wt (+op')	Lána et al. (2010)
R-85	Fish (Large size)		whole	Mississippi River (Upper)	2004-2005	USA		11.16 ng/g wet wt	Blocksom et al. (2010)
R-86	Fish (Large size)		whole	Missouri River	2004-2005	USA		8.18 ng/g wet wt	Blocksom et al. (2010)
R-87	Fish (Large size)		whole	Ohio River	2004-2005	USA		18.32 ng/g wet wt	Blocksom et al. (2010)
R-88	Fish (Small size)		whole	Mississippi River (Upper)	2004-2005	USA		6.57 ng/g wet wt	Blocksom et al. (2010)

No.	Species	n <sup>a</sup>	Analyte	River	Year	Country	T-HCH <sup>b</sup>	T-DDT <sup>c</sup>	References
R-89	Fish (Small size)		whole	Missouri River	2004-2005	USA		5.47 ng/g wet wt	Blocksom et al. (2010)
R-90	Fish (Small size)		whole	Ohio River	2004-2005	USA		15.60 ng/g wet wt	Blocksom et al. (2010)
R-91	European eel	30	Muscle	Garigliano River (Campania region)	2005-2006	Italy		52.91 ng/g wet wt	Ferrante et al. (2010)
R-92	Brown trout ( <i>Salmo trutta</i> )	9	Muscle	Quemquentreu river	2006	Argentina		1.7 ng/g wet wt	Ondarza et al. (2011)
R-93	Brown trout ( <i>Salmo trutta</i> )	9	Liver	Quemquentreu river	2006	Argentina		7.4 ng/g wet wt	Ondarza et al. (2011)
R-94	Sábalo fish ( <i>Prochilodus lineatus</i> )	7	Muscle	Río de la Plata basin	2003-2004	Argentina	9 ng/g wet wt	340 ng/g wet wt (+op')	Colombo et al. (2011)
R-95	Eel	10	Muscle	Rivers in South Canterbury	2009	New Zealand		33.5 ng/g wet wt *	Stewart et al. (2011)
R-96	Brown trout	5	Muscle	Rivers in South Canterbury	2009	New Zealand		16.8 ng/g wet wt *	Stewart et al. (2011)
R-97	Largemouth bass		Muscle	Blackwater River (Lower)	2004	USA (Florida)		ND	Karouna-Renier et al. (2011)
R-98	Largemouth bass		Muscle	Perdido River (Lower)	2004	USA (Florida)		0.51 ng/g wet wt	Karouna-Renier et al. (2011)
R-99	Largemouth bass		Muscle	Yellow River (Lower)	2004	USA (Florida)		ND	Karouna-Renier et al. (2011)
R-100	Tilapia	8	Muscle	Noha River (Okinawa-Manko)	2006	Japan	24 ng/g fat wt	3800 ng/g fat wt	Malarvannan et al. (2011)
R-101	Tilapia	8	Muscle	Hija River (Okinawa-Kadena)	2006	Japan	4.7 ng/g fat wt	1100 ng/g fat wt	Malarvannan et al. (2011)
R-102	Tilapia	8	Muscle	Shikaza River (Okinawa-Onna village)	2005	Japan	10 ng/g fat wt	680 ng/g fat wt	Malarvannan et al. (2011)

<sup>a</sup> No. of analyzed samples; Mean (n≥2) or single determination values (n=1) are listed for PCB, T-HCH and T-DDT (\* Median value)

<sup>b</sup> T-HCH =  $\alpha$ -HCH +  $\beta$ -HCH +  $\gamma$ -HCH

<sup>c</sup> T-DDT = pp'-DDE + pp'-DDD + pp'-DDT

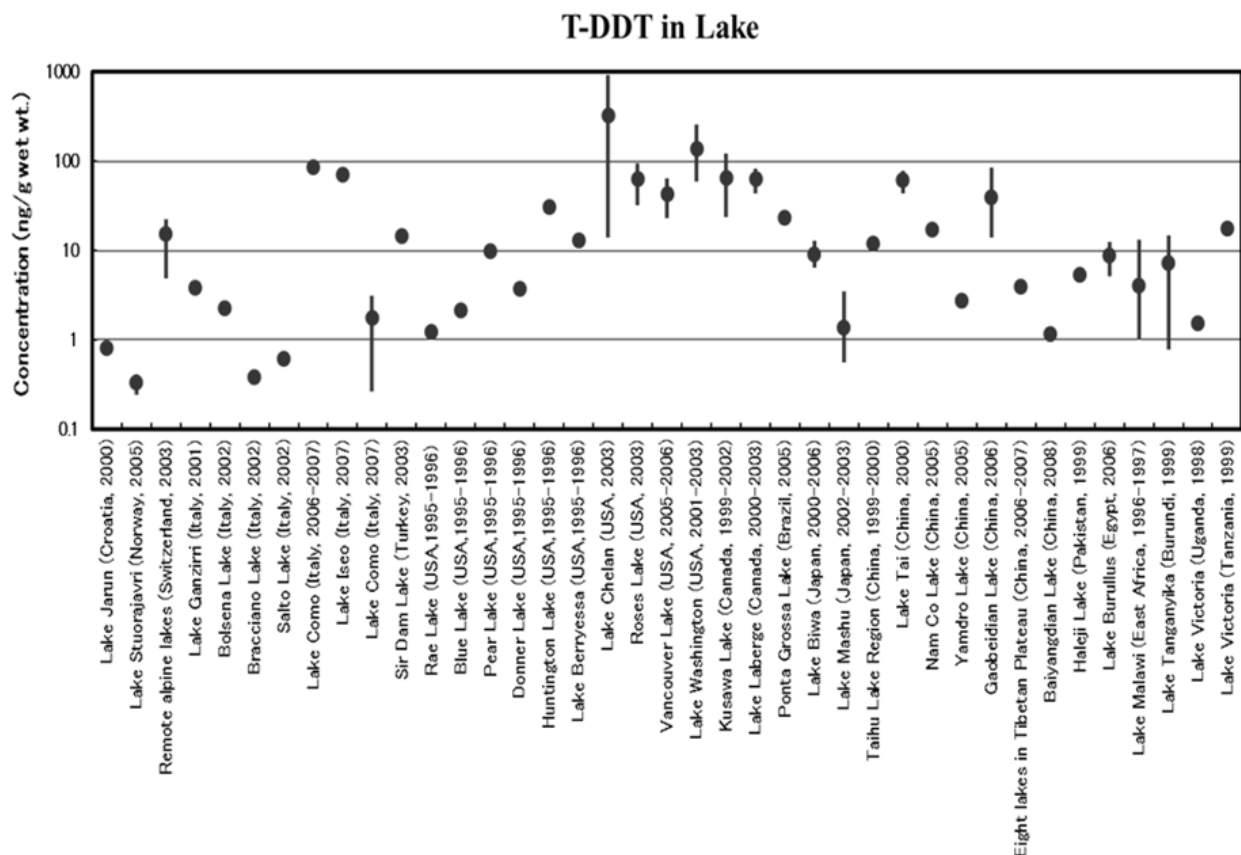
**Table 2.** Concentrations of T-HCH and T-DDT in fish from rivers in the world

Residue of T-DDT and T-HCH in fish from lakes in the world is shown in Figures 1 and 2, respectively, for the concentration data (ng/g wet wt.) and (ng/g fat wt. and ng/g dry wt. possible to be calculated as ng/g wet wt.). Each of the T-DDT and T-HCH concentration data was compared for the 38 lakes surveyed in 8 countries of Europe and America and 8 countries of Asia and Africa from 1995 to 2008. Each data is shown as single determination value (n=1) and mean (n≥2) or mean and range values (n≥2) for the surveys in plural fish species, sampling sites and survey years.

T-DDT concentrations in the fish from the lakes of America were relatively high and the higher concentrations were detected in USA like the previous report [102]. Those of Europe were relatively low except for two lakes in Italy. Those of Asia and Africa including Japan were relatively low as a whole, although relatively high concentrations were detected in a

part of lakes in China. T-HCH concentrations in the fish from the lakes in the world were relatively low. The highest T-HCH concentration was 46 ng/g wet wt. in Taihu Lake of China [46] and relatively low concentration of 1.4 ng/g wet wt. (average,  $n=7$ ) was detected in Lake Biwa of Japan [22-28].

Residue of T-DDT and T-HCH in fish from rivers in the world is shown in Figures 3 and 4, respectively, for the concentration data (ng/g wet wt.) and (ng/g fat wt. and ng/g dry wt. possible to be calculated as ng/g wet wt.). Each of the T-DDT and T-HCH concentration data was compared for the 28 rivers surveyed in 8 countries of Europe and America, 4 countries of Asia, Africa and Oceania from 2000 to 2009. Each data is shown as single determination value ( $n=1$ ) and mean ( $n\geq 2$ ) or mean and range values ( $n\geq 2$ ) for the surveys in plural fish species, sampling sites and survey years.



**Figure 1.** Residue of T-DDT in fish from lakes in the world



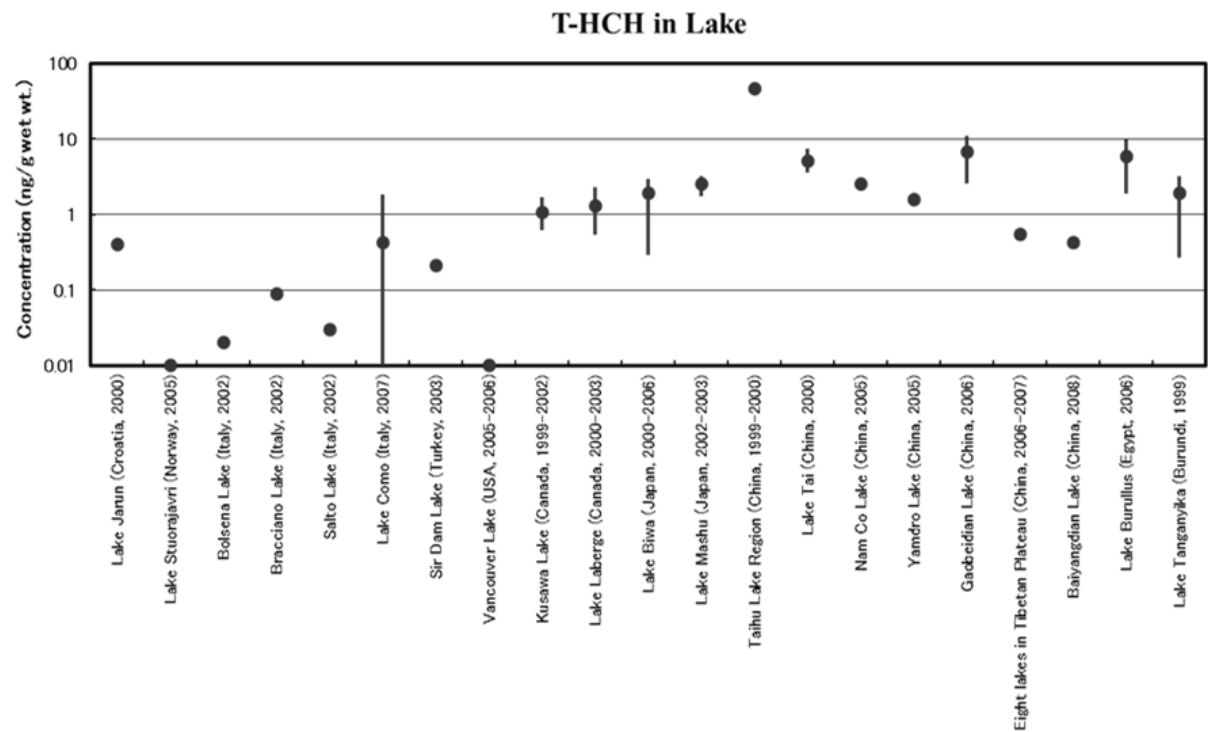


Figure 2. Residue of T-HCH in fish from lakes in the world

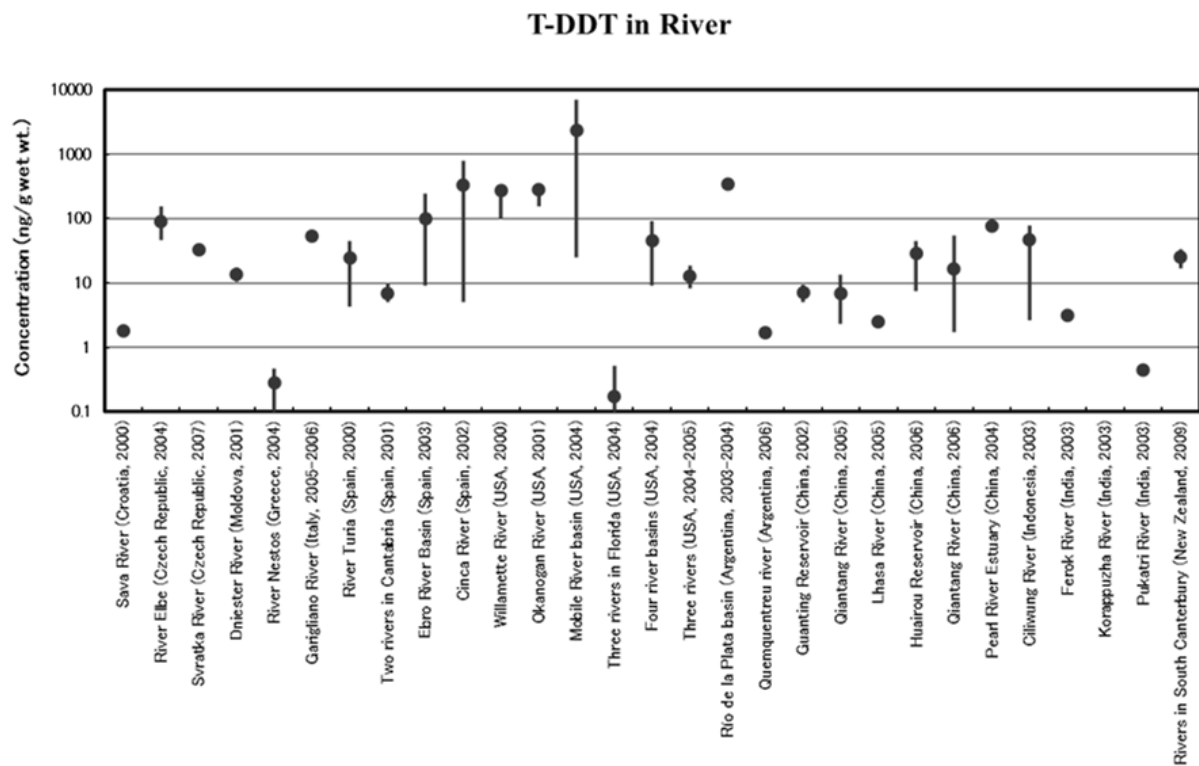
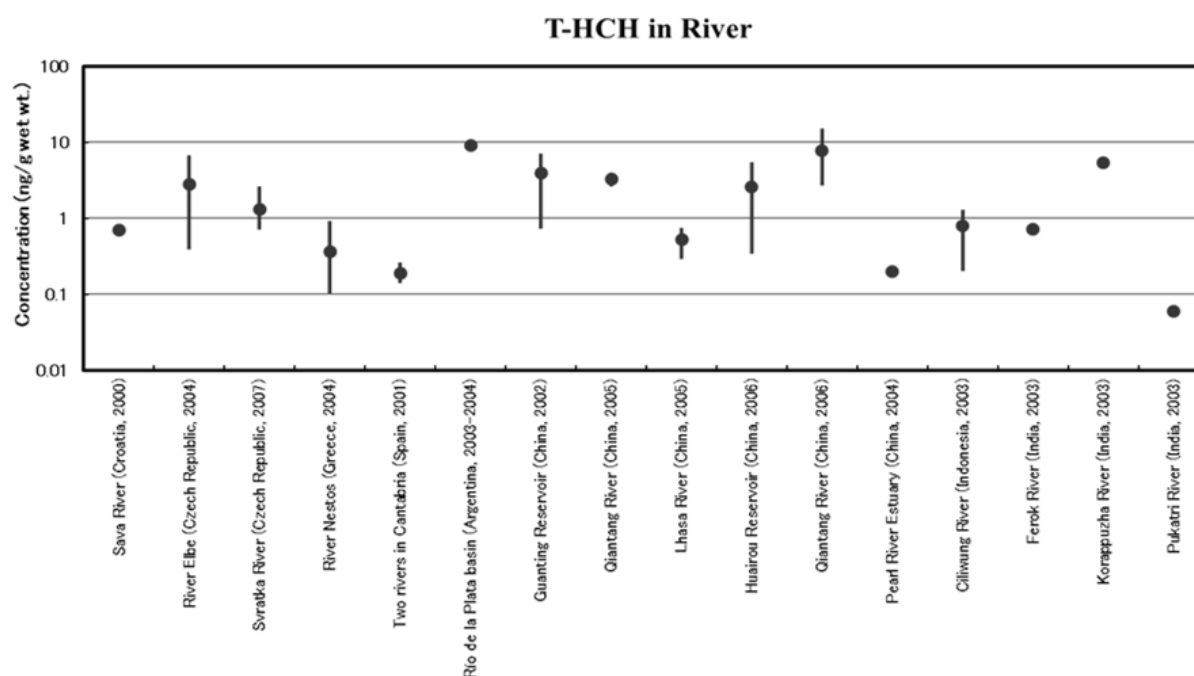


Figure 3. Residue of T-DDT in fish from rivers in the world





**Figure 4.** Residue of T-HCH in fish from rivers in the world

T-DDT concentrations in the fish from the rivers of USA were relatively high like the previous report [102]. Those of Europe were relatively low except for a part of lakes in Czech Republic and Spain. Those of Asia and Africa were relatively low as a whole, although relatively high concentrations were detected in a part of rivers of China.

T-HCH concentrations in the fish from the rivers in the world were relatively low like the lakes in the world and the highest T-HCH concentration was 7.7 ng/g wet wt. (average,  $n=8$ ) in Qiantang River of China [101].

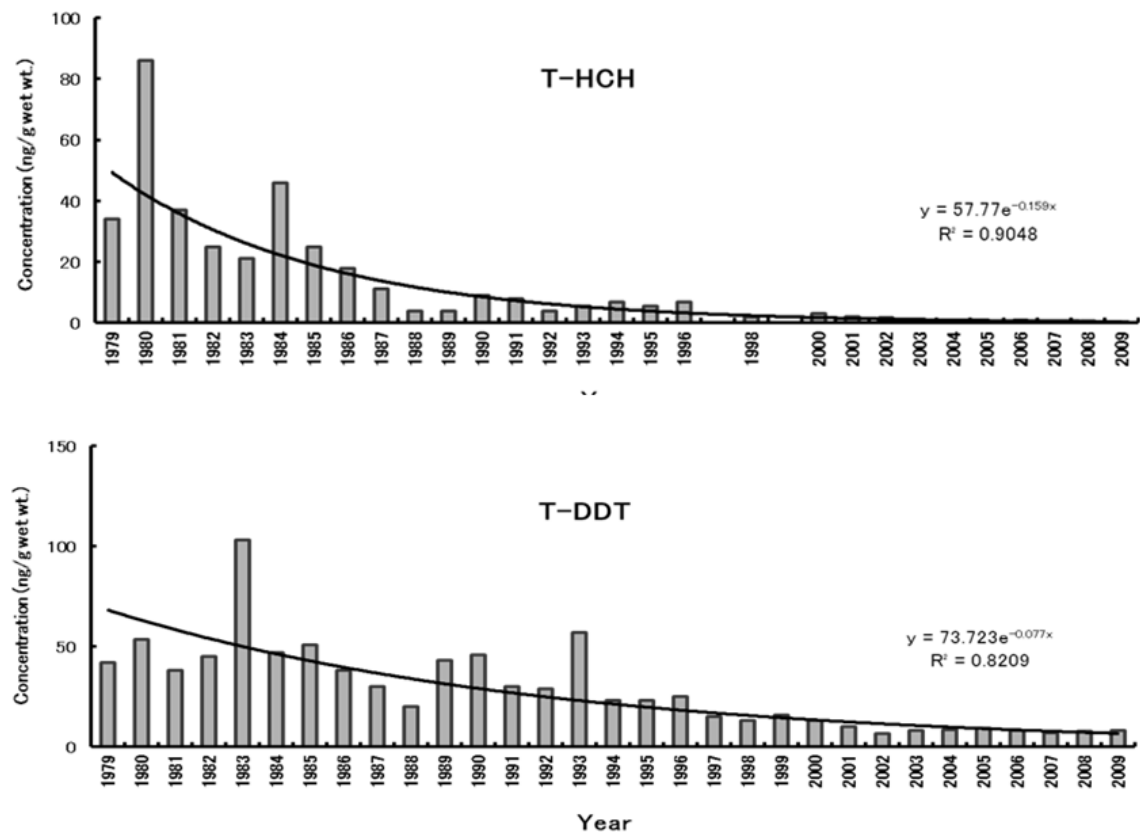
### 3. Long-term trends of T-DDT and T-HCH in fish from lakes in the world

T-HCH and T-DDT concentrations in fish were calculated as the simple sum of the constituents. T-HCH is the sum of  $\alpha$ -HCH,  $\beta$ -HCH and  $\gamma$ -HCH and T-DDT is the sum of pp'-DDT, pp'-DDD and pp'-DDE. For Japan, T-DDT and T-HCH concentration data in Japanese dace from Lake Biwa were cited from reports of Ministry of the Environment, Japan (1980 – 2011) [1-31]. For Canada and USA, T-DDT concentration data in lake trout from Lake Ontario and Lake Michigan were cited from a report of Environment Canada and U.S. Environmental Protection Agency (2007) [32] and a source figure of U.S. Environmental Protection Agency (2009) [33], respectively. For Sweden, T-DDT concentration data in pike from Lake Storvindeln were cited from a source figure of Swedish EPA (2002) [34].

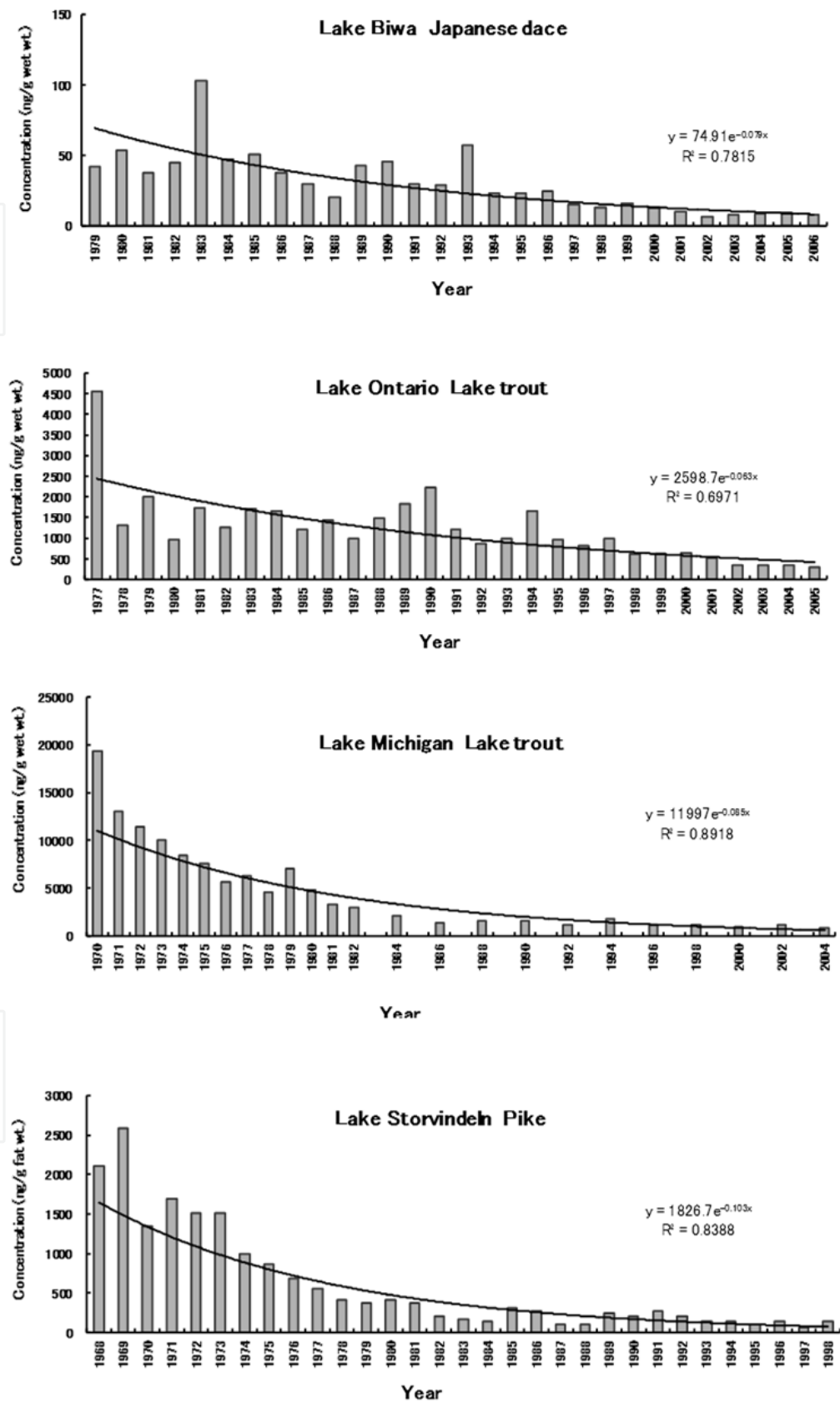
All data were analyzed using Microsoft Excel graph wizard. For each location and analyte, nonlinear procedure was used to fit the exponential model:  $y=ae^{-kx}$ , where  $y$  is the concentration in each composite sample,  $x$  is the sampling date, and  $a$  and  $k$  are model parameters estimated by nonlinear procedure to obtain a specific model that best fits the data.

Long-term trends of T-DDT and T-HCH in Japanese dace from Lake Biwa are shown from 1979 to 2009 in Figure 5 along with long-term trends estimated using the first-order model. Both insecticides were found to decline in a consistent pattern. Model parameters (a, k) and R<sup>2</sup> values were calculated for each of the data sets and are shown in Figure 5. Half-lives (t<sub>1/2</sub>) were calculated for T-DDT and T-HCH from a parameter (k). The t<sub>1/2</sub> values were 9 years for T-DDT and 4 years for T-HCH.

Long-term trends of T-DDT in fish from Lake Biwa, Lake Ontario, Lake Michigan and Lake Storvindeln are similarly shown in Figure 6. The t<sub>1/2</sub> values of T-DDT were 9, 11, 8 and 7 years, respectively, in Lake Biwa, Lake Ontario, Lake Michigan and Lake Storvindeln. There were no wide differences in the t<sub>1/2</sub> values of T-DDT in the fish among the four lakes. The same extent of the decline rate in the T-DDT inflow into the four lakes was presumed from the same extent of the decline rate of T-DDT in the fish of the four lakes.



**Figure 5.** Long-term trends of T-DDT and T-HCH in Japanese dace from Lake Biwa



**Figure 6.** Long-term trends of T-DDT in fish from Lake Biwa, Lake Ontario, Lake Michigan and Lake Störvindeln

#### 4. Composition of T-DDT and T-HCH in fish from lakes and rivers in the world

Composition of T-DDT in fish from lakes in the world is shown in Figure 7 for the survey data in the 25 lakes in 15 countries of Europe, America, Asia and Africa from 1996 to 2008 [22-28, 35, 39, 41, 42, 44, 47, 49-53, 55, 56, 58, 60, 61, 64, 65, 67, 68, 70]. Few (ND~0.6 %) or low (1~6 %) percentage of *PP'*-DDT was detected in Lake Biwa and Lake Mashu of Japan, Lake Tai and three lakes of China, Lake Stuorajavri of Norway, Lake Ganzirri of Italy and Sir Dam Lake of Turkey. Metabolites of *PP'*-DDT (*pp'*-DDE and *pp'*-DDD) were detected at high percentage and long-term no use of DDT was presumed in the countries. On the other hand, high percentage (44~88 %) of *PP'*-DDT was detected in Lake Edward of Uganda, three lakes of Ghana, Lake Burullus of Egypt, Lake Victoria of Tanzania and Ponta Grossa Lake of Brazil. DDT was presumed to be used in the countries in recent years or in the sampling date. The use of organochlorine pesticides such as DDT was prohibited or restricted in the 1970's for Japan and 1980's for Europe and in 1983 for China. This corresponded well to the survey data described above. Low percentage of *PP'*-DDT in the fish from Lake Michigan and Lake Superior of USA was reported in the previous report [102]. The percentage of *PP'*-DDT was similarly low (5 %) in the fish from Vancouver Lake in the present report. In Africa, the use of DDT was restricted in the 1980's and low percentage of *PP'*-DDT was reported in the fish from Manzara Lake or Idku Lake in the previous report [102]. The present result in Lake Burullus was different from the previous report and the use of DDT was presumed in recent years or in the sampling date. Details of the restriction on the use of DDT in Egypt were obscure.

Composition of T-DDT in fish from rivers in the world is shown in Figure 8 for the survey data in the 16 rivers in 8 countries of Europe, America, Asia, Africa and Oceania from 2000 to 2009 [51, 68, 73, 77, 79, 80, 83, 86, 88, 91, 94, 95, 98, 100, 101].

In China, relatively high percentage (38 %) of *PP'*-DDT was detected in Qiantang River of China (2005). However, low percentage (2~10 %) of *PP'*-DDT was detected in Lhasa River, Huairou Reservoir and Qiantang River (2006). In Japan, India, Greece, Spain and USA, low percentage (ND~12 %) of *PP'*-DDT was similarly detected. No use of DDT was presumed in recent years or in the sampling date in all rivers except for Qiantang River of China surveyed in 2005.

Composition of T-HCH in fish from lakes in the world is shown in Figure 9 for the survey data in the 16 lakes in 8 countries of Europe, America, Asia and Africa from 1996 to 2008 [22-28, 35, 42, 44, 51, 53, 56, 58, 60, 61, 65, 68]. Composition of T-HC in Lake Biwa of Japan ( $\alpha$ -HCH 7 %,  $\beta$ -HCH 91 % and  $\gamma$ -HCH 3 %) was similar to that in Lake Tai of China ( $\alpha$ -HCH 10 %,  $\beta$ -HCH 84 % and  $\gamma$ -HCH 6 %). It was known that technical HCH ( $\alpha$ -HCH 65~70 %,  $\beta$ -HCH 6~14 %,  $\gamma$ -HCH 10~13 % and  $\delta$ -HCH 5~8 %) had been used in China, India and former Soviet Union since 1979 [103]. In Japan, technical HCH was also used without purification until 1971. This is the reason for the similarity of HCH composition in the fish between Japan and China (Lake Biwa and Lake Tai). However, the percentage of  $\beta$ -HCH in Yamdro Lake and Gaobeidian Lake was relatively low and the percentage of  $\beta$ -HCH in Lake

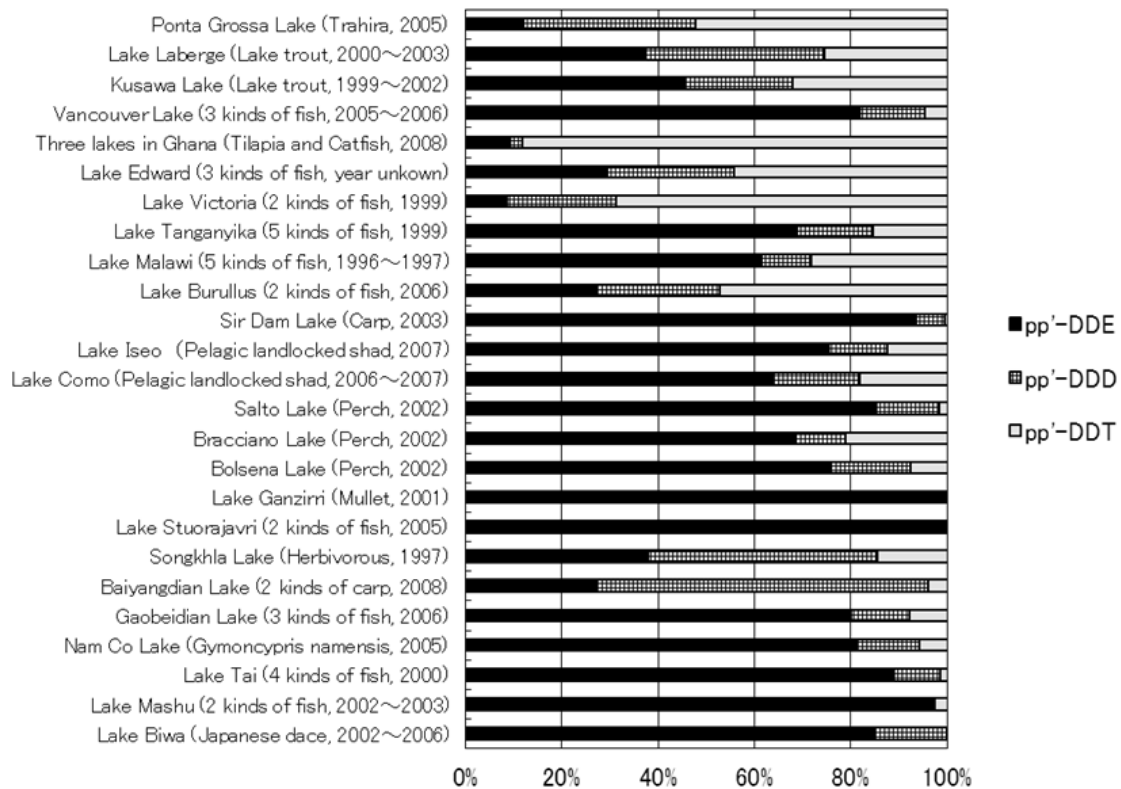


Figure 7. Composition of T-DDT in fish from lakes in the world

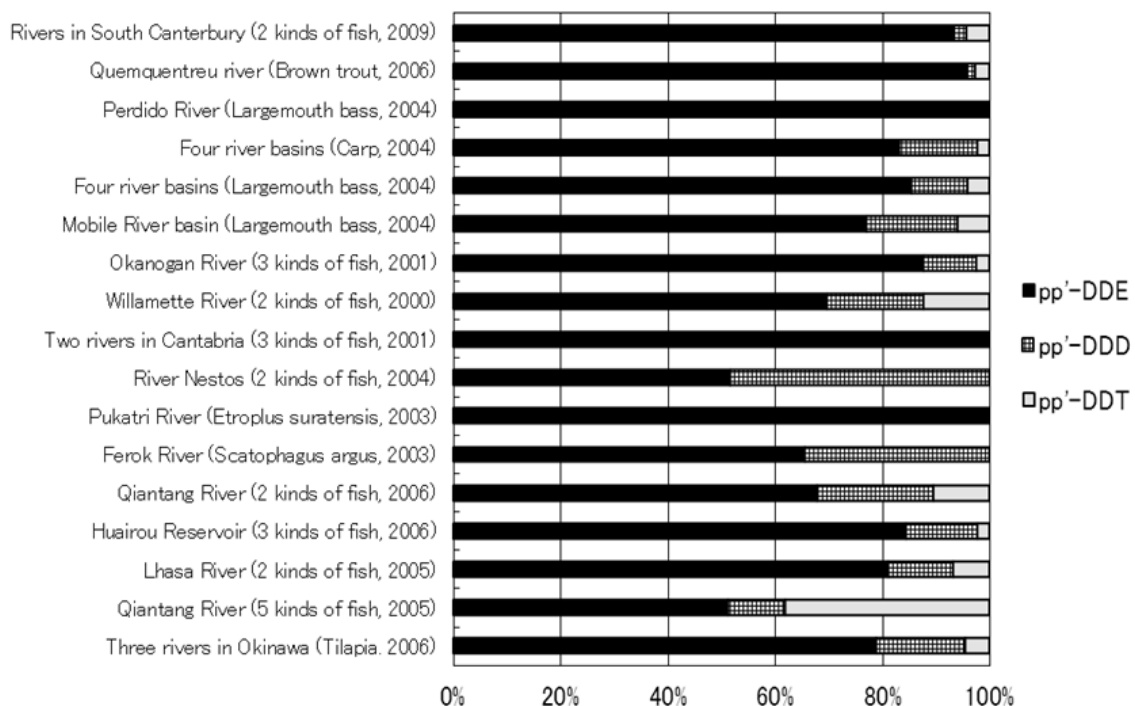
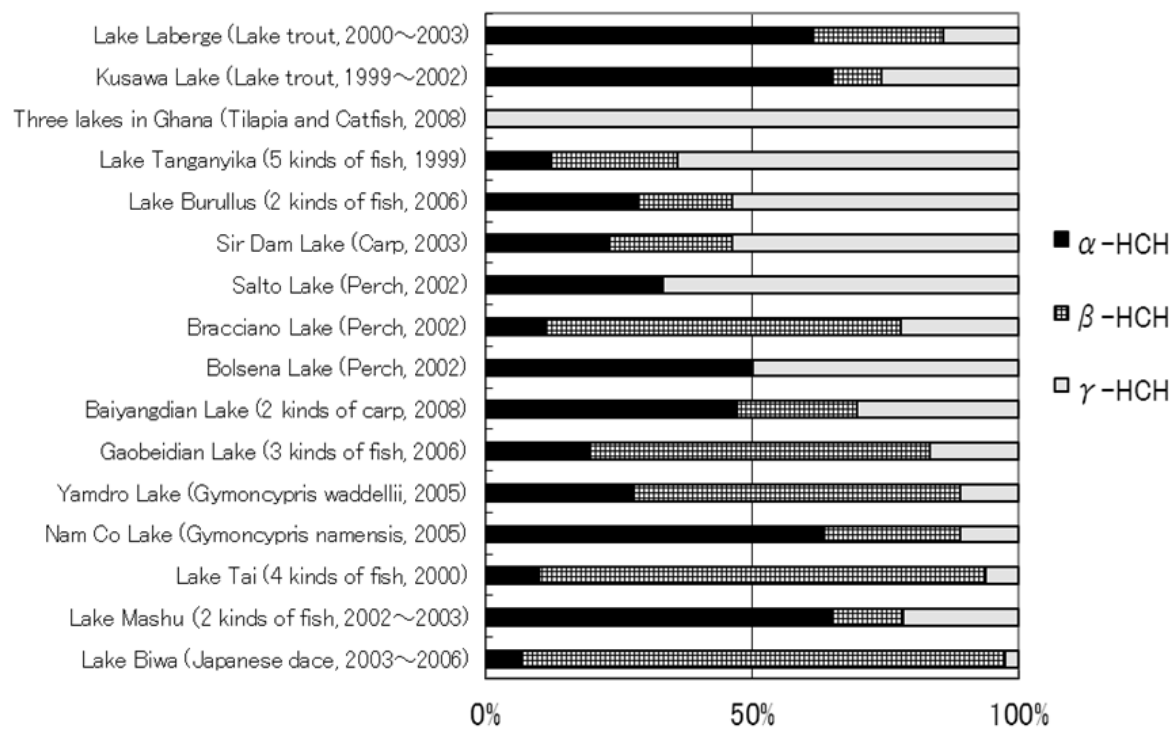


Figure 8. Composition of T-DDT in fish from rivers in the world



**Figure 9.** Composition of T-HCH in fish from lakes in the world

Biwa of Japan was higher than that in the lakes of China. There was a probability of high  $\beta$ -HCH percentage in Japan because  $\beta$ -HCH was highly persistent and the period after prohibition on the use of  $\beta$ -HCH was longer in Japan than in China. On the other hand, the percentage of  $\beta$ -HCH in Rainbow trout and Japanese dace from Lake Mashu in Japan was low. Composition of T-HC in Japanese dace from Lake Mashu ( $\alpha$ -HCH 64~67 %,  $\beta$ -HCH 11 % and  $\gamma$ -HCH 22~25 %) was much different from that of Lake Biwa ( $\alpha$ -HCH 6~8 %,  $\beta$ -HCH 89~92 % and  $\gamma$ -HCH 2~3 %). This is probably because HCH was loaded in Lake Mashu through the atmosphere [104] and the percentage of  $\alpha$ -HCH in the water of Lake Mashu was much higher than that of Lake Biwa.

On the other hand, purified lindane ( $\gamma$ -HCH: more than 99 %) was used in Europe and America differently from Japan, China, etc. This is probably because of the high percentage of  $\gamma$ -HCH in Bolsena Lake and Salto Lake of Italy, Sir Dam Lake of Turkey, Lake Burullus of Egypt, Lake Tanganyika of Burundi and three lakes of Ghana.

Composition of T-HCH in fish from rivers in the world is shown in Figure 10 for the survey data in the 11 rivers in 5 countries of Europe and Asia from 2001 to 2006 [51, 68, 73, 77, 86, 92, 100, 101].

In China, high percentage of  $\gamma$ -HCH was detected in Qiantang River and Huairou Reservoir differently from the survey data in the lakes. The use of lindane was presumed in the two river basins. In Korappuzha River of India, the use of technical HCH was presumed and this corresponded well to the report that technical HCH had been used in China, India and former Soviet Union since 1979 [103]. Particularly high percentage of  $\gamma$ -HCH was detected in Nestos River of Greece and this corresponded well to the use of lindane in Europe and

America similarly in the case of the lake. For Okinawa Prefecture in Japan, the use of technical HCH in Shikaza River and Hija River and the use of lindane in Noha River were presumed from the composition of T-HCH shown in Figure 10. The high percentage of  $\gamma$ -HCH in Noha River did not correspond to the use of technical HCH in Japan.

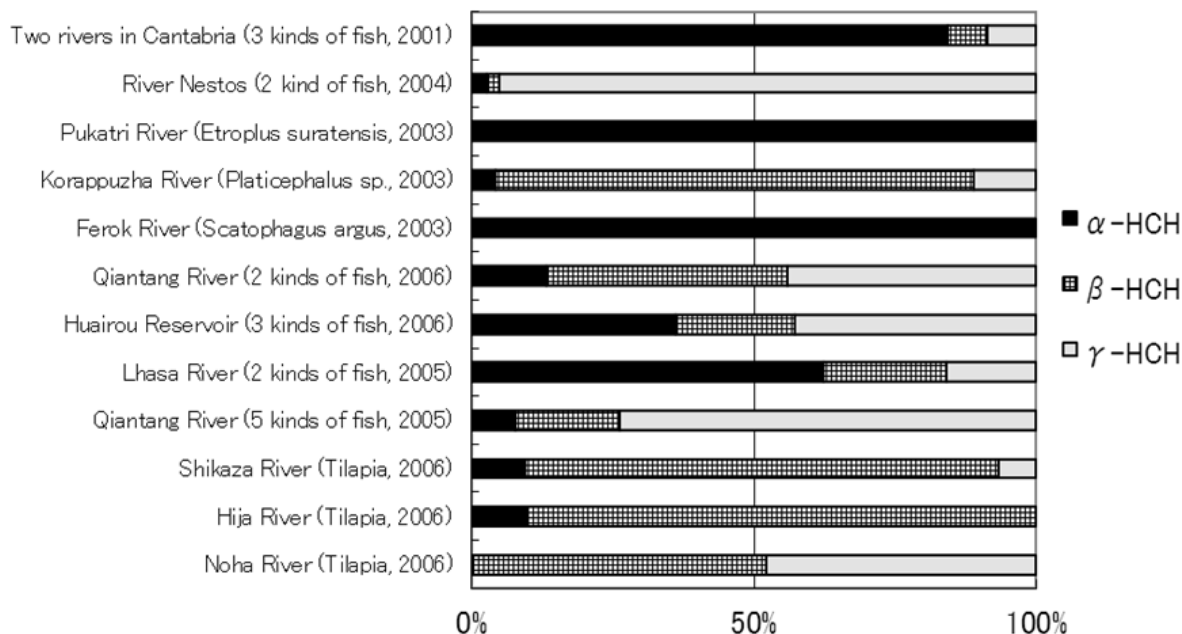


Figure 10. Composition of T-HCH in fish from rivers in the world

## 5. Conclusion

T-DDT concentrations in the fish from the lakes and rivers of America were relatively high, but those of Europe, Asia and Africa were relatively low. T-HCH concentrations in the fish were relatively low in both of the lakes and rivers in all over the world. T-DDT and T-HCH compositions were respectively compared among lakes and rivers from America, Europe, Asia and Africa. DDT was presumed to be used in Uganda, Egypt, Tanzania and Brazil in recent years or in the sampling date from the high percentage of *pp'*-DDT in the composition of T-DDT and its metabolites in the several kinds of fish from Lake Edward, Lake Burullus, Lake Victoria and Ponta Grossa Lake. No use of DDT was presumed in USA and European countries from the low percentage of *pp'*-DDT in the lake fish in the countries. Technical HCH was presumed to be used in Japan, China and India from the low percentage of  $\gamma$ -HCH in the composition of T-HCH in the lake and the river fish in the countries. On the contrary, Lindane was presumed to be used in the countries of Europe and Africa from the high percentage of  $\gamma$ -HCH in the lake and the river fish in the countries. Half-lives ( $t_{1/2}$ ) of T-DDT in fish from lakes in Japan, Canada, USA and Sweden were calculated from the long-term monitoring data using an exponential decay model to evaluate the decline rate of DDT contamination in the lake environment. The  $t_{1/2}$  values were 9 years for Lake Biwa in Japan, 11 years for Lake Ontario in Canada, 8 years for Lake Michigan in USA and 7 years for Lake Störvindeln in Sweden. There were no wide differences in the  $t_{1/2}$  values of T-DDT in the fish among the four lakes.



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