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Butyltin Compounds in River Otters (*Lutra canadensis*) from the Northwestern United States

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Abstract. Butyltin compounds, including mono-, di-, and tributyltin (MBT, DBT, and TBT) were measured in livers of 40 adult river otters (*Lutra canadensis*) collected from rivers and coastal bays in Washington and Oregon, USA. Butyltins were found in all the river otters, at a concentration range of 8.5–2,610 ng/g, WW. The greatest concentration of total butyltins of 2,610 ng/g, WW, was found in a river otter collected in Puget Sound from Fort Ward, Washington. River otters collected near areas with major shipping activities, such as the Puget Sound, contained significantly greater concentrations (geometric mean: 367 ng/g, WW) of butyltins than those from rivers. Among butyltin compounds, MBT and DBT predominated in livers. The concentrations of butyltins in river otters ranged from comparable (Puget Sound) to less (rivers) than what was found in coastal cetaceans.

Tributyltin (TBT) has been used for almost 30 years as an antifouling agent in marine paint formulations to prevent the accumulation of barnacles and slime on boat hulls. During the early to middle 1980s, concern about the environmental impact of TBT and its degradation products, mono- (MBT) and dibutyltin (DBT), grew due to the ability of TBT to cause deleterious effects in a variety of aquatic organisms at very low water concentrations. For instance, growth and reproduction in the most sensitive species—notably stenoglossan gastropods and oysters—can be impaired by TBT at water concentrations of only a few parts per trillion (ng/L) (Bryan *et al.* 1986; Alzieu *et al.* 1990). The estimated tissue concentration of TBT in the rock shell *Thais clavigera* and *T. bronni* to cause imposex has been 10–20 ng/g, WW (Horiguchi *et al.* 1994). Based on the results of many laboratory studies documenting sublethal and lethal effects of TBT on a variety of marine organisms, many countries (including the United States) passed legislation limiting the use of TBT-based antifouling paints in boats in the late 1980s (Uhler *et al.* 1993; Fent 1996). Water concentrations of butyltins (BTs = MBT + DBT + TBT) decreased in most locations subsequent to the regulations (Valkirs *et al.* 1991; Fent 1996). However, in certain locations, TBT concentrations in water exceeded the guidelines or were cause for concern even after several years of being banned (Dowson *et al.* 1992; Ritsema 1994; Fent and Hunn 1995; Chau *et al.* 1997). Further, many researchers have noted little or no reduction in TBT concentrations in sediments several years after regulations were enacted (Quevauviller *et al.* 1994; Dowson *et al.* 1994; Fent and Hunn 1995; Fent 1996). Similarly, no apparent decline in TBT concentrations have been observed in large harbors that handle ships legally painted with TBT-containing antifouling paints (Fent 1996; Chau *et al.* 1997). Because of its continued use on large ships and apparent persistence in sediment, TBT is still a concern in near-shore urban waterways (Meador 1997).

Despite the significance of TBT contamination in coastal waters, studies conducted until the mid-1990s have monitored TBT accumulation in a relatively small number of species, principally oysters, dogwhelks, and mussels. For a complete assessment of the impact of butyltin compounds in aquatic ecosystems, it is necessary to examine the accumulation and effects of these compounds in various organisms in the food chain. Our recent studies have shown that butyltin compounds accumulate in body tissues of marine mammals and water birds at significant concentrations, and that butyltin contamination in higher trophic level animals in the food chain is widespread (Iwata *et al.* 1995; Kannan *et al.* 1996, 1997a, 1998; Kannan and Falandysz 1997; Kim *et al.* 1996a, 1996b; Guruge *et al.* 1996; Tanabe *et al.* 1998).

In contrast to the marine environment, data on the occurrence of BTs in fresh water including rivers, are scarce. Butyltin contamination in lakes and rivers originates from the use of TBT as an antifouling agent as well as the use of DBT and MBT as stabilizers in polyvinylchloride (PVC) pipes (Maguire *et al.* 1982; Fent and Hunn 1995; Stäb *et al.* 1996; Kannan *et al.* 1997b).

The river otter is a top predator of most aquatic food chains that has adapted to a wide variety of aquatic habitats, from marine environments to high mountain lakes of North America (Toweill and Tabor 1982). Otters exhibit differing degrees of sociality and spacing based on habitat, shelter availability, and food abundance (Reid *et al.* 1993). Otter home ranges (approximately 11 river miles) are largely defined by local topography

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and overlap extensively within and among sexes, exhibiting varying degrees of mutual avoidance and tolerance depending on seasonal dispersion and availability of food and shelter (Reid *et al.* 1993). However, otters do maintain territories within home ranges that are delineated by scent marking and latrine sites. Areas within territories are used almost exclusively by the defending otter, excluding otters of the same sex (*i.e.*, female otters exclude other females and family groups while males exclude other males). Otters eat a wide variety of fish species and aquatic invertebrates such as crabs, crayfish, and mussels. This makes otters good integrators of their aquatic environments and a useful surrogate species for determining both wildlife and human chemical exposure and potential harmful effects.

In this study, concentrations of butyltins in the liver of North American river otters collected from various rivers and coastal estuaries from Oregon and Washington were determined to elucidate contaminant concentrations and to delineate sources.

Materials and Methods

Samples

Otter samples used in this study are part of a larger U.S. Geological Survey project (SIS 70-67) designed to identify contaminants most responsible for observed reproductive organ hypoplasia in young male river otters from the lower Columbia River. During the 1996–97 trapping season, frozen river otter carcasses were obtained from licensed trappers throughout Oregon and Washington. The use of river otter carcasses taken by trappers utilizes a resource that would normally have been discarded and reduces the need for removal of additional animals. A subset of 40 adult river otters (38 males and 2 females) purchased from trappers in 1996–97 were selected for this study. Nearly equal numbers (9 to 12 under each category) were obtained from the Puget Sound (heavy ship use), bays and small rivers (some ship and boat use), large rivers (ship and boat use), and small creeks, small rivers, or lakes (small boat or no boat use). Frozen otter carcasses were necropsied at the Oregon State University Veterinary Diagnostics Laboratory (OSU-VDL), basic morphometric information was recorded, and tissue collection was made for chemistry and histopathology. Frozen carcasses were allowed to thaw prior to necropsy. Body condition, weights, and measurements were recorded. A portion of the liver was placed in chemically clean jars and frozen at −20°C. Liver samples were shipped on dry ice to the Aquatic Toxicology Laboratory at Michigan State University for butyltin analysis. River otters were aged by counting cementum layers (each layer or annuli represents 1 year) of an extracted canine tooth (Matson 1981) at the Matson Laboratory, Milltown, Montana. Sample details are given in Table 1 and sampling locations are shown in Figure 1.

Chemical Analysis

The analytical method used for the determination of MBT, DBT, and TBT in otter tissues has been described elsewhere (Iwata *et al.* 1995; Kannan *et al.* 1995a, 1995b). Briefly, acidified tissue samples were homogenized with 70 ml of 0.1% tropolone-acetone, and the solvent was transferred to 100 ml of 0.1% tropolone-benzene. Moisture in the organic extract was removed using 35 g anhydrous sodium sulfate and then the concentrated extract was propylated with *n*-propyl magnesium bromide (about 2 mol/L in THF solution, Tokyo Chemical Industries, Portland, OR) as a Grignard reagent. The derivatized extract was passed through a 6 g florisil-packed wet column for clean-up. The eluate from the Florisil column was rotary-evaporated and injected into a gas chromatograph.

For the quantification of butyltin compounds, a capillary gas chromatograph with a flame photometric detector (GC-FPD) was used. The flame photometer was operated using a hydrogen-air-nitrogen flame and was equipped with a 610-nm bandpass filter that is selective for tin-containing compounds.

One hundred nanograms each of butyltin trichloride, dibutyltin dichloride, and tributyltin chloride were spiked into the liver of lake trout (*Salvelinus namaycush*), containing butyltins below the limit of detection, passed through the whole analytical procedure and used as an external standard. Only freshly derivatized external standards prepared along with every set of eight samples were used to estimate concentrations. Concentrations were quantified by comparing peak heights of butyltins in samples with those in the external standards. Tributylhexyltin (synthesized by the reaction of *n*-hexyl magnesium bromide with tributyltin chloride) was added to each sample as an internal standard prior to extraction. A procedural blank was analyzed with every set of eight samples to check for interfering compounds and to correct sample values, if necessary. Monobutyltin, probably originating from commercial solvents or reagents that came into contact with PVC containing this compound as a stabilizer, was found at trace levels in reagent blanks. The values obtained for MBT in samples were therefore corrected for blank concentrations. However, blanks never contained TBT. The detection limits of MBT, DBT, and TBT were 7, 2.4, and 1 ng/g WW respectively. The average recovery rates of monobutyltin trichloride, dibutyltin dichloride, and tributyltin chloride dissolved in hexane, spiked into the sample matrix, and passed through the whole analytical procedure were between 90 and 110% for each compounds. All concentrations reported in this study refer to butyltin species as the corresponding ion, and they were not corrected for the recovery of the internal standard (which was between 85 and 105%).

Results and Discussion

Butyltin compounds were found in the liver of all river otter tissues analyzed (Table 1). Hepatic concentrations of total BTs in otters collected from various locations in Oregon ranged from 8.5 to 102 ng/g, WW, whereas those from Washington ranged from 11 to 2,610 ng/g, WW.

Concentrations of BTs varied depending on the location of sampling. The greatest BT concentration of 2,610 ng/g, WW, was found in a river otter collected in Puget Sound from Fort Ward, Washington. Nevertheless, additional samples collected from Fort Ward contained 25-fold lesser concentrations than the river otter that had the greatest concentration. BT concentrations of $> 1{,}000$ ng/g, WW, were found in individual otters collected from Camano Island, Bremerton, and Eglon. All these locations are a part of the Puget Sound in Washington and have shipping activities (Table 1). The variation in BT contamination in individual otters collected within a geographic area may be due to the movement of otters and their dietary habits. Large numbers of animals need to be analyzed from these locations with elevated concentrations to refer to specific reasons for individual variations in concentrations.

In order to evaluate the sources of butyltins, animals were grouped into four categories based on the collection location as: Puget Sound, Large Rivers, Small Rivers–Bays, and Small Rivers (Table 1). Major shipping activity is located in the Puget Sound, whereas in large rivers little shipping activity, but considerable small craft activity and sewage effluent are expected (river otters from the Willamette River of Oregon were not trapped in the lower portion of the river, where

Table 1. Concentrations (ng/g, ww) of butyltin compounds in the liver of adult male river otters collected in 1996–97 from Oregon and Washington, USA[a]

Sample ID	Location	Age (years)	Body Weight (kg)	Body Length (cm)	MBT	DBT	TBT	BTs[b]	Large Rivers	Small Rivers	Bay	Puget Sound
Oregon Sites												
RAG-001	Yaquina River (Toledo)	4	8.18	121	23	26	3.6	53		•	•	
RAG-065	Yaquina River (Toledo)	5	8.39	109	21	77	3.8	102		•	•	
RAG-230	Yaquina River (Toledo)	2	7.48	111	<7	20	2.8	23		•	•	
RAG-026	Willamette River (Albany, RM 108)	4	9.07	121	30	34	2.7	67	•			
RAG-027	Willamette River (Albany, RM 108)	4	8.84	117	29	10	1.9	41	•			
RAG-028	Willamette River (Albany, RM 108)	4	7.71	118	13	15	2.1	30	•			
RAG-030	Willamette River (Albany, RM 116)	4	9.53	118	18	34	1.8	54	•			
RAG-058	Willamette River (Eugene, RM 175)	4	7.71	113	40	40	4.6	85	•			
RAG-066	Willamette River (Albany, RM 118)	5	8.39	121	57	10	3.6	71	•			
RAG-067	Willamette River (Albany, RM 118)	1	9.07	123	15	11	1.6	28	•			
RAG-075	Willamette River (Salem, RM 79)	3	8.16	107	18	29	3.5	51	•			
RAG-029	Columbia River (Scappoose, RM 88)	3	9.53	113	19	50	2.7	72	•			
RAG-022	Nehalem River (Mist)	2	7.26	112	7.1	13	<1	20		•		
RAG-041	Nehalem River (Mist)	6	9.75	120	27	30	1.5	59		•		
RAG-208	Nehalem River (Mist)	2	8.16	118	<7	8.5	<1	8.5		•		
RAG-215	Humbug Creek (Vinemaple)	8	7.94	115	<7	16	1.4	17		•		
Washington Sites												
RAG-092[c]	Camano Island	6	10.4	123	1,200	490	12	1,700				•
RAG-126	Soleduck River (Forks)	3	9.53	117	21	56	2	79		•		
RAG-137	Soleduck River (Forks)	4	8.16	113	8.2	17	1.1	26		•		
RAG-148	Fort Ward (Bainbridge Island)	7	9.07	120	1,540	1,050	16	2,610				•
RAG-192	Fort Ward (Bainbridge Island)	3	8.85	115	40	100	2.3	142				•
RAG-194	Fort Ward (Bainbridge Island)	2	8.16	114	35	65	<1	100				•
RAG-195	Fort Ward (Bainbridge Island)	8	9.98	119	27	72	1.4	100				•
RAG-157	Gig Harbor	2	10.9	124	290	470	35	795				•
RAG-193	Bremerton	6	10.2	120	590	610	21	1,220				•
RAG-200	Petroleum Creek	6	8.85	115	14	36	2.9	53		•		
RAG-221	Petroleum Creek	8	9.53	118	16	33	3.9	53		•		
RAG-232[c]	Petroleum Creek	2	6.8	110	<7	30	1.3	31		•		
RAG-202	Puyallup River, Tacoma	3	9.53	121	24	100	2.7	127		•		•
RAG-176	Willapa Bay (North River)	3	6.35	105	22	17	<1	39		•	•	
RAG-209	Willapa Bay (Nemah River)	6	7.26	113	<7	8.5	1.5	12		•	•	
RAG-252	Willapa Bay (Willapa River)	2	7.94	114	78	72	8.4	158		•	•	
RAG-257	Willapa Bay (North River)	6	8.85	119	14	32	1.5	48		•	•	
RAG-213	Silverdale	2	8.16	106	20	55	5.3	82				•
RAG-237	Silverdale	3	7.94	117	38	410	9.2	457				•
RAG-214	Eglon	2	5.9	108	24	102	2	128				•
RAG-240	Eglon	5	9.75	124	120	1460	16	1600				•
RAG-217	Forks (Wentworth Lake)	5	7.94	104	<7	8.8	1.7	11		•[d]		
RAG-218	Sequim (Meadowbrook Creek)	9	9.3	116	<7	16	1.7	18		•	•	
RAG-236	Grays Harbor (Hoquiam River)	5	8.39	114	22	82	4.7	109		•	•	

[a] Samples are grouped according to their habitat type as: Puget Sound, large rivers, small rivers, and Bay
[b] MBT + DBT + TBT
[c] Two female otters were included
[d] One otter trapped at a lake

shipping activity and repair occurs). Sewage effluent in the large river category would come from cities like Eugene, Corvallis, Albany, and Salem on the Willamette River. Small rivers–bays are expected to have ocean fishing boat activity, moderate to light small craft activity, and reduced sewage effluent. The small river category would have the least amount of sewage effluent and small craft activity. Significantly higher concentrations were found in river otters collected in the Puget Sound, whereas the concentrations were the least in river otters from small rivers (Table 2). This suggests that the otters inhabiting areas near boating activity have been exposed to elevated concentrations of butyltins. High accumulation of butyltins in otters from the Puget Sound (a marine area) could be influenced by greater bioavailability of TBT in sea water than in fresh water. In waters of pH 8 and above, butyltins, particularly TBT, exist as hydroxides or chlorides (neutral forms), whereas in waters of lower pH, the cation is the predominant form (Fent and Looser 1995). The neutral forms are more readily accumulated in aquatic biota than the cation. Therefore the presence of sources (boating) in combination

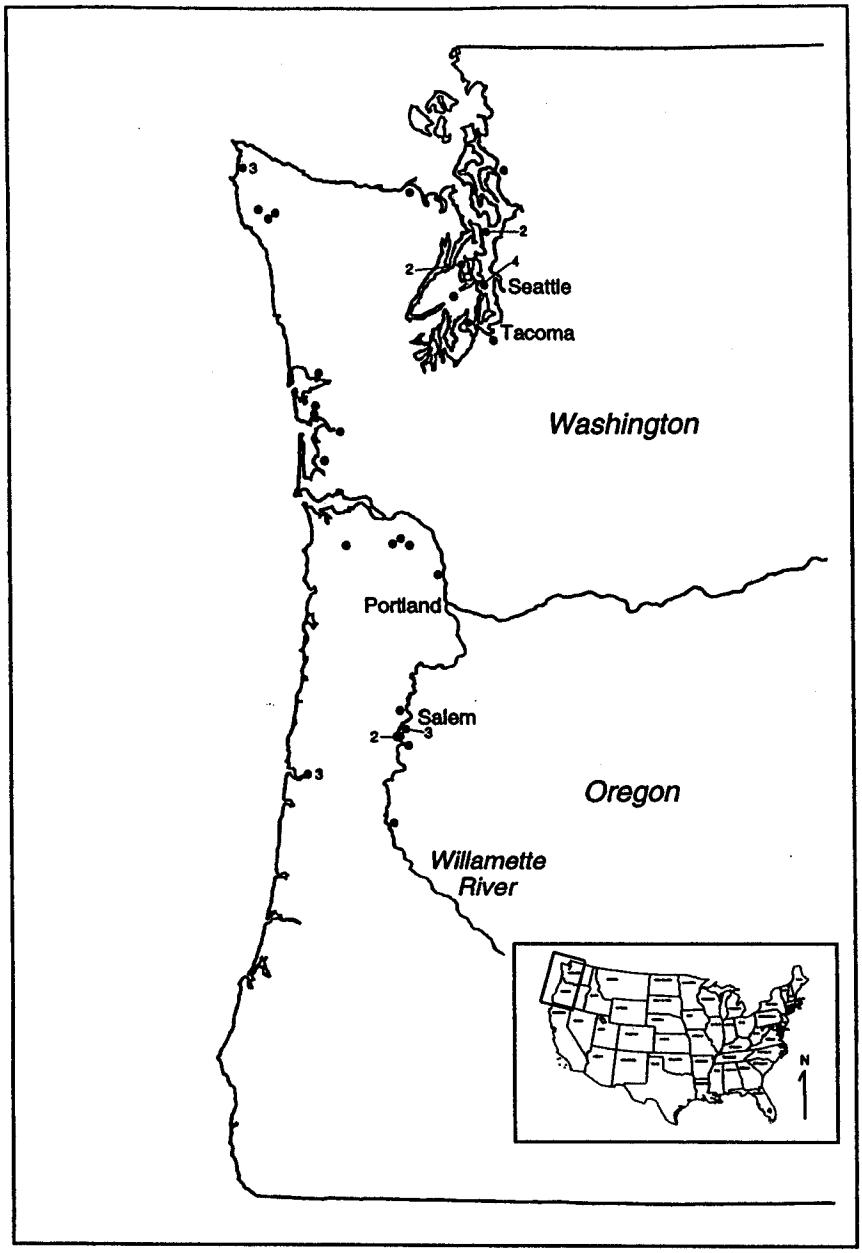

Fig. 1. Map of Washington and Oregon showing sampling locations (dots) of river otters along the Willamette River and in the Puget Sound. The values adjacent to the dots represent the number of otters analyzed from the location

with greater bioavailability of TBT in sea water have contributed to higher concentrations in Puget Sound otters than those from rivers.

A few studies have reported butyltin contamination in coastal sediments, bivalve mollusks, and fish from Washington and Oregon. Concentrations of total butyltins in bivalve mollusks collected from Grays Harbor and Willapa Bay, Washington, during 1989–90, were 100 and 4 ng/g, WW, respectively, whereas those from the Columbia River and Yaquina Bay, Oregon, were 94 and 56 ng/g, WW, respectively (Uhler *et al.* 1993). Butyltin concentrations in fish livers from the Elliott Bay in Washington were 290–380 ng/g, WW, whereas those from the Coos Bay were 690 ng/g, WW (Krone *et al.* 1996). Abandonment of oyster growing areas or oyster shell thickening and deformation have been reported in the Coos Bay,

Table 2. Geometric mean mono- (MBT), di- (DBT), and tributyltin (TBT), and sum of butyltins (BTs) residue concentrations (ng/g, ww) in livers of adult river otters in Puget Sound, large rivers, small rivers–bays, and small rivers aquatic habitat types

Group	p Value	Puget Sound	Large Rivers	Small Rivers–Bays	Small Rivers
MBT	0.0001	96.2^{A}	23.7^{B}	13.0^{B}	8.0^{B}
DBT	0.0001	231.1^{A}	21.8^{B}	29.6^{B}	20.9^{B}
TBT	0.0086	5.6^{A}	2.6^{AB}	2.4^{AB}	1.4^{B}
BTs	0.0001	367.3^{A}	51.7^{B}	47.8^{B}	31.3^{B}

One-way ANOVA, general linear models procedure, Tukey's studentized range test, Alpha = 0.05. Rows of these categories sharing same letter are not significantly different

Table 3. Comparison of percentages (%) of TBT and MBT + DBT in total butyltin concentrations (BTs) in the livers of marine mammals collected from various locations

Species	(MBT + DBT)/BTs	TBT/BTs	Reference
Bottlenose dolphin, coastal Florida, USA	93	7	Kannan *et al.* (1997a)
Atlantic spotted dolphin, coastal Florida, USA	87	13	Kannan *et al.* (1997a)
Southern sea otters, coastal California, USA	55	45	Kannan *et al.* (1998)
Bottlenose dolphin, coastal Italy	80	20	Kannan *et al.* (1996)
Risso's dolphin, Pacific coast of Japan	77	23	Kim *et al.* (1996a)
Finless porpoise, Ise Bay, Japan	75	25	Iwata *et al.* (1995)
Gingkgo-toothed beaked whale, Japan Sea	77	23	Tanabe *et al.* (1998)
Largha seal, Hokkaido coast, Japan	90	10	Tanabe *et al.* (1998)
Dall's porpoise, Sanriku coast, Japan	70	30	Tanabe *et al.* (1998)
Baird's beaked whale, Japanese coast	89	11	Tanabe *et al.* (1998)
Bottlenose dolphin, off Taiji, Japan	83	17	Tanabe *et al.* (1998)
Killer whale, off Taiji, Japan	93	7	Tanabe *et al.* (1998)
Ribbon seal, Hokkaido coast, Japan	64	36	Tanabe *et al.* (1998)
Northern fur seal, Hokkaido coast, Japan	71	29	Tanabe *et al.* (1998)
River otters, Washington and Oregon			
Puget Sound	98.5	1.5	Present study
Large rivers	95	5	Present study
Small rivers–bays	95	5	Present study
Small rivers	95.5	4.5	Present study

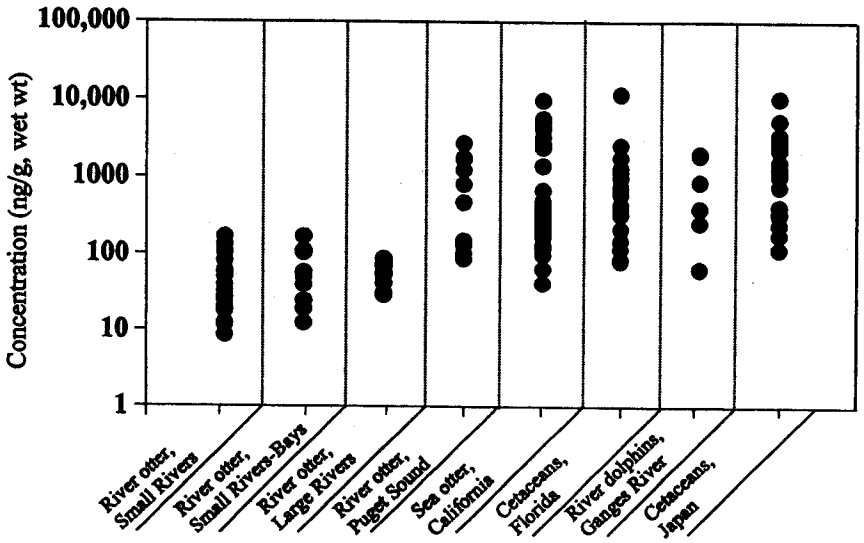

Fig. 2. Comparison of butyltin concentrations in river otters from Washington and Oregon (present study) with those of cetaceans from other regions. Data for sea otters, cetaceans from Florida, Ganges River dolphins, and cetaceans from Japan were cited from Kannan *et al.* (1998), Kannan *et al.* (1997a), Kannan *et al.* (1997b), and Tanabe *et al.* (1998), respectively

Orgeon (Wolniakowski *et al.* 1987). While these results suggested butyltin contamination in coastal regions, studies on butyltins in rivers in the northwestern United States are scarce. Our results suggest the occurrence of butyltin compounds in rivers, although at relatively lower concentrations than in the Puget Sound.

Among various butyltin compounds, DBT and MBT were the predominant compounds in river otters (Table 2). DBT contributed to greater proportions of total BT concentrations in the livers of marine mammals from several coastal locations, which was explained by the metabolic transformation of TBT to DBT and MBT in aquatic organisms (Lee 1991). The composition of TBT in total BTs was 1.5% for those animals from Puget Sound, 5% from large rivers, 5% from small rivers–bays, and 4.5% from small rivers (Table 3). This proportion of TBT in total BT concentrations in river otters was less than those found in marine mammals from several coastal locations, which contained 7–45% of the total butyltins as TBT (Table 3). Efficient metabolic transformation of TBT to MBT and DBT and exposure to butyltin sources with greater proportions of MBT and DBT could explain the reason for the abundance of MBT and DBT in river otters. Exposure of otters to butyltin sources originating from the leaching of MBT and DBT, which are used as stabilizers in PVC pipes (Fent and Müller 1991; Chau *et al.* 1992), could be a possible explanation. In fact, all of the Willamette River otters were taken from 100 to 230 km upstream from navigable waters. Age- and size-related variation in butyltin concentrations in river otters could not be examined due to the small number of samples of individual locations.

The butyltin concentrations measured in river otters' from Puget Sound were comparable to those reported for cetaceans

from coastal areas in the United States and Japan (Figure 2). Nevertheless, butyltin concentrations in otters collected from rivers were less than those of marine mammals from coastal areas. Most river otters are found in freshwater environs, and those associated with marine environs probably spend a portion of their time in freshwater streams. Therefore, river otters were not expected to have body burdens of butyltins at levels reported in other coastal marine mammals. Moreover, fur-bearing animals such as river otters may eliminate butyltin compounds during their annual molting cycle since butyltins have a tendency to bind to structural proteins in fur and hair. Elimination of butyltin compounds by molting has been reported in Stellar sea lion, a fur-bearing marine mammal (Kim *et al.* 1996b). In any case, our results suggest the accumulation of butyltin compounds in river otters from inland rivers.

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