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Acute Toxicity of Waterborne Se(IV), Se(VI), Sb(III), and Sb(V) on Red Seabream (*Pargus major*)

K. Takayanagi

National Research Institute of Fisheries Science 6-31-1 Nagai, Yokosuka 238-0316, Japan

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Both selenium (Se) and antimony (Sb) are toxic and have been listed as priority pollutants by the US Environmental Protection Agency (US EPA). Substantial quantities of Se and Sb enter aquatic ecosystems from both natural and anthropogenic sources. Anthropogenic sources account for about 41 x 10^6 kg/yr and 18 x 10^6 kg/yr of Se and Sb, respectively (Nriagu and Pacyna, 1988). The major anthropogenic sources of these elements are fossil fuel burning and industrial waste from metal smelters and steel plants (Nriagu and Pacyna, 1988). The ultimate sink for Se and Sb is the marine environment. Although, the toxicity of Se to freshwater organisms has been extensively studied (US EPA, 1987), toxicities of Se and Sb to marine organisms are not well examined.

Both elements can exist in several oxidation states in seawater. Three oxidation states of Se in oxic seawater, Se(-II), Se(IV), and Se(VI), have been reported (Cutter and Bruland, 1984; Takayanagi and Wong, 1985), while two oxidation states of Sb in oxic seawater, Sb(III) and Sb(V), have been detected (Bertine and Lee, 1983; Middleburg et al., 1988). As toxicities of these elements may be different depending on their oxidation state, it is necessary to examine the toxicity of each chemical form separately to assess its environmental effects on the marine ecosystem. Here we report acute toxicity of Se(IV), Se(VI), Sb(III), and Sb(V) to red seabream, *Pargus major*, an important commercial saltwater fish.

MATERIALS AND METHODS

Juvenile *Pargus major* were obtained from Shizuoka Prefectural Field Station (Hamaoka, Shizuoka). They were spawned, hatched out artificially, and fed on a formula feed (Kyowa Hakko Co., Tokyo). Feeding was stopped two days before the experiments. Initial body weights are shown in Table 1.

Present address: National Research Institute of Aquaculture, 422-1 Nakatsuhamaura, Nansei, Mie 516-0193, Japan

Test solutions were made by adding a small amount of a stock solution containing Se(IV), Se(VI), Sb(III), or Sb(V) to seawater. The stock solution with Se(IV) was made with $NaSeO_3$, that with Se(VI) with $NaSeO_4$, that with Sb(III) with $SbCl_3$, and solutions with Sb(V) were made with $SbCl_5$ and with $K[Sb(OH)_6]$. All the reagents used were analytical grade. The seawater used was first passed through sand and activated-charcoal filters. The salinity of seawater was 33.7 ppt.

Acute toxicity tests were conducted in 30-L glass aquaria according to the procedures recommended by American Society for Testing and Materials (1998). A group of 8 *P. major* (about 3 month old) was transferred to each aquarium and exposed to a range of concentrations, as shown in Table 1, for up to 96 hours under static conditions at 20±1 °C. The Se and Sb concentrations of the test solutions were verified at the beginning and at the end of the experiment by determining the actual concentrations using the hydride-generation atomic-absorption method (Grasshoff et al., 1983). The pH was also determined every day. In order to maintain an adequate oxygen level, each aquarium was aerated. Mortality of test fish was recorded after 24, 48, 72, and 96 hr exposure. Median lethal concentration (LC_{50}) values were estimated by probit analysis. No mortality occurred in the control.

Table 1. Experimental conditions for the Se and Sb acute toxicity tests

Group	Body Weight* of *P. major* (g)	Test Solution	Range of Concentration Tested (mg/L, as Se or Sb)	pH Range of Test Solution
1	2.05±0.14	Control	-	7.9 - 8.2
2	2.04±0.11	HCl	-	5.0
3	2.03±0.09	Se (IV): $NaSeO_3$	5.4 - 18.2	7.8 - 8.1
4	2.05±0.10	Se (VI): $NaSeO_4$	40 - 137	7.8 - 8.1
5	2.74±0.16	Sb (III): $SbCl_3$	7.8 - 25.7	4.9 - 7.8
6	2.94±0.15	Sb (V): $SbCl_5$	0.40 - 1.06	7.8 - 8.1
7	2.88±0.18	Sb (V):	2.8 - 10.3	7.8 - 8.1

*: mean ± S.D.

RESULTS AND DISCUSSION

The acute toxicities of Se(IV), Se(VI), Sb(III), and Sb(V) found for *P. major* are listed in Table 2. As expected, LC_{50} progressively decreased as the duration of exposure increased. The exception was Sb(V). The LC_{50}'s were constant over 96 hrs: 0.93 mg/L using $SbCl_5$ and 6.9 mg/L using $K[Sb(OH)_6]$. For the $SbCl_3$ test, a decrease in the pH of the test solution was found (Table 1). Therefore, a low pH seawater was prepared with HCl for use as a control for the $SbCl_3$ test in order to assess pH effects. All the test fish survived in the HCl-adjusted seawater. Therefore, pH was a negligible factor, and the mortality found in the $SbCl_3$ dilution waters was considered to have been caused by the $SbCl_3$.

Table 2 LC_{50} values (mg/L, as Se or Sb) of the Se and Sb species for *P. major*

	Se (IV) $NaSeO_3$	Se (VI) $NaSeO_4$	Sb (III) $SbCl_3$	Sb (V) $SbCl_5$	Sb (V) $K[Sb(OH)_6]$
$24hrLC_{50}$	15.0	>137	15.5	0.93	6.9
$48hrLC_{50}$	12.2	>137	15.5	0.93	6.9
$72hrLC_{50}$	12.0	107	15.2	0.93	6.9
$96hrLC_{50}$	11.5	76	12.4	0.93	6.9

The different oxidation states of each element caused diverse effects on the test fish. *P. major* appeared to be more sensitive to Se(IV) than to Se(VI). Se(IV), the lower oxidation state, was about seven times more toxic than Se(VI). The 96-hr LC_{50} was 11.5 mg/L for Se(IV), but 76 mg/L for Se(VI). Most of the published data on the acute toxicity to fish of Se is for the Se(IV) form. Furthermore, only a few data exist for saltwater (or saltwater acclimatize) fish. Some of the published data are included in Table 3. Reported 96-hr LC_{50} values for Se(IV) range from 0.6 mg/ L on haddock (*Melanogrammus aeglefinus*) larva to 32.5 mg/L on coho salmon (*Oncorhynchus kisutch*) fry. On the other hand, for Se(VI) they range from 9.8 mg/L on striped bass (*Worone saxatilis*) prolarva to 149 mg/L on chinook salmon (*Oncorhynchus tschawytscha*) fry. Hamilton and Buhl (1990) compared the relative sensitivity of chinook salmon and coho salmon to the two oxidation states of Se, and they reported that Se(IV) was more toxic (Table 3). Niimi and LaHam (1976) also reported that Se(IV) was 4 to 10 times more toxic than Se(VI) to the freshwater species they studied, newly hatched zebrafish (*Brachydanio rerio*). Our results, which found Se(IV) to be more toxic than Se(VI), are consistent with those reported results.

P. major was more sensitive to Sb(V) than to Sb(III). The 96-hr LC_{50}'s were 12.4 mg/L, 0.93 mg/L, and 6.9 mg/L for Sb(III), Sb(V) using $SbCl_5$, and Sb(V) using $K[Sb(OH)_6]$, respectively. In contrast to Se, for Sb, the higher oxidation state, Sb(V), was more toxic than the lower oxidation state, Sb(III). Even for Sb(V), the different test chemicals yielded a significant difference in the degree of acute toxicity. $SbCl_5$ was seven times more toxic than $K[Sb(OH)_6]$. Published results on the acute toxicity of Sb are even scarce. Heitmuller et al (1981) reported the 96-hr LC_{50} of Sb (test chemical is not specified) to sheepshead minnows (*Cyrinodan variegatus*) to be 6.2 to 8.3 mg/L (Table 3). Our results on Sb(III) and Sb(V) using $K[Sb(OH)_6]$ are comparable, but our $SbCl_5$ test yielded a much lower 96-hr LC_{50} value. For the freshwater species, Lin and Hwang (1998) reported the 96-hr LC_{50} of $SbCl_5$ to be 35.5 mg/L for larval tilapia (*Oreochromis mossambicus*). This large difference is difficult to explain. It may be just due to a difference between the physiology of saltwater fish and freshwater fish.

Table 3 96hrLC_{50} values (mg/L, as Se or Sb) of Se and Sb to salt and brackish water fish

	Se (IV)		Se (VI)	Sb	Sb (III)	Sb (V)	
	$NaSeO_3$	H_2SeO_3	$NaSeO_4$		$SbCl_3$	$SbCl_5$	$K[Sb(OH)_6]$
Pargus major[1] (juvenile)	10.6		70		12.9	0.93	6.9
Worone saxatilis[2] (prolarvae)			9.8				
Worone saxatilis[2] (juvenile)			85.8				
Worone saxatilis[3] (63 day old)	1.6						
Oncorhynchus tschawytscha (fry)[4]	23.4		149				
Oncorhynchus kisutch[4] (fry)	32.5		39.0				
Melanogrammus aeglefinus[2] (larva)		0.6					
Cyrinodan variegatus[2] (juvenile)		6.7					
Cyrinodan variegatus[3] (juvenile)				6.2-8.3			
Wenidia menidia[2] (juvenile)		9.7					
Lagodon rhomboides[2] (juvenile)	4.4						
Paralichthys dentatus[2] (embryo)		3.5					
Pseudopleuronecies omericanus (larva)[2]		14.2					

1: this study, 2: US EPA (1987), 3:Palawski et al. (1985), 4: Hamilton & Buhl (1990), 5: Heitmuller et al (1981)

It may not be easy to compare our acute toxicity data with other published work because the test chemicals and the life stages of the test organisms were different, but it is clear that Se(IV) is more toxic than Se(VI), while Sb(V) is more toxic than Sb(III). In order to establish water quality criteria for these elements, it is necessary to consider their chemical forms.

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