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Aluminum in lake water and organs of a fish *Tribolodon hakonensis* in strongly acidic lakes with a high aluminum concentration

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Abstract Aluminum in lake water and in the organs of the fish *Tribolodon hakonensis* was investigated in Lake Usoriko (pH 3.6), Lake Inawashiroko (pH 5.0), and the Tenryu River (pH 7.7). The concentration of total soluble aluminum in the water was 0.51 $mg\,l^{-1}$ in Usoriko, 0.05 $mg\,l^{-1}$ in Inawashiroko, and less than 0.01 $mg\,l^{-1}$ in the Tenryu. The chemical forms of soluble aluminum in the acid water were characterized as Al^{3+}, AlL^{2+}, and $AlL^{\leqq 1+}$. More than 90% of soluble aluminum in the water of Usoriko was Al^{3+}, whereas AlL^{2+} was dominant in the water of Inawashiroko. The aluminum concentration in the organs of *T. hakonensis* in Usoriko was 42 $\mu g\,g^{-1}$ wet weight in gills, 4.2 $\mu g\,g^{-1}$ in muscle, 6.9 $\mu g\,g^{-1}$ in bone, 12.7 $\mu g\,g^{-1}$ in liver, 6.0 $\mu g\,g^{-1}$ in kidney, and 6.0 $\mu g\,g^{-1}$ in intestine, indicating accumulation of aluminum in the gills. The aluminum concentration in the organs of *T. hakonensis* living in Inawashiroko was approximately the same, in spite of the difference in water chemistry of the two acid lakes, especially for pH and aluminum. This suggests that aluminum accumulation might be controlled in the fish living in the acid lakes. In contrast, the aluminum concentration in the gills of *T. hakonensis* from the Tenryu was 2 $\mu g\,g^{-1}$.

Key words Acid lake · Fish · Aluminum · *Tribolodon hakonensis*

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Introduction

The water of Lake Usoriko is a mixture of acidic waters of volcanic origin supplied from springs of about pH 2 and a stream of pH 4.7, and neutral water inflows from the catchment area.

The biota of Usoriko is classified as acidophile or acid tolerant, being composed of algae, aquatic vascular plants, moss, zooplankton, insects, fish, and birds, which form a stable and characteristic ecosystem. Fossil records of shoots of the moss *Drepanocladus fluitans* with attached diatoms *Pinnularia braunii* var. *amphicephala* and *Nitzschia palea* as indicator plants of an acidic environment show that Usoriko was also strongly acidic in ancient times. The ^{14}C data for the cones of the terrestrial tree *Picea glehnii* discovered in the paleo lake sediment with *D. fluitans* show that the acid lake dates back at least 30 000 years.

Among the organisms living in the ecosystem of Lake Usoriko, the fish *Tribolodon hakonensis* is noteworthy, because it lays its eggs in neutral water, and the fry, which are acid sensitive, then migrate to the acidic lake. The fish grow to adulthood by feeding on plankton and insects, and the adult fish return to the inflowing neutral river to breed (Satake et al. 1995). This is a typical form of ecological adaptation for survival, because the acid water of the lake is fatal to most other species of fish, especially in the egg and fry stages (Ikuta et al. 1999). One of the reasons for the acid tolerance of adult *T. hakonensis* is the presence of chloride cells, which may adjust the concentration of H^+ inside the body relative to that outside (Mashiko et al. 1973). However, the distribution of *T. hakonensis* in nonacidic environments in the Japanese islands, the Korean peninsula, and Russia is also worth considering to understand the distribution of *T. hakonensis* in acidic environments. In many cases, *T. hakonensis* found in these areas migrates between fresh water and seawater. The combination of neutral inflow streams and acidic lakes where *T. hakonensis* migrates is analogous to the combination of fresh water and seawater. In both cases, the regulation of ions by the fish is common in waters of different quality. In the case of the combination of neutral water and acidic water, the fish, especially in the stages of egg and fry, can probably escape from high proton and aluminum concentrations by such migration. A high aluminum concentration is usually toxic to fish and could be an additional problem for survival in strongly acidic water.

Acidification of the environment causes an increase in the concentration of aluminum in the aquatic environment, where bioavailable aluminum is suspected to have a detrimental effect on fish (Cronan and Schofield 1979; Baker and Schofield 1982). The toxicity of aluminum is linked to its biological availability, but the processes by which aluminum is adsorbed into, excreted from, or deposited in the body are still poorly understood. To understand the behavior of aluminum and its toxicity to fish in acidic environments, it is important to study its distribution in the body and to clarify whether it accumulates in any particular organ.

In this study, we collected and analyzed the lake water and organs of *T. hakonensis* in acidic environments and also a neutral environment for comparison, and considered the reasons for the difference in aluminum concentration between the lake water and the organs, and for the aluminum tolerance of *T. hakonensis.*

Sampling sites and methods

Sampling sites

Samples of water were collected from Lake Usoriko with acidic hot springs located at the northern part of Usoriko (41°19′N, 141°05′E) and the inflowing neutral stream Maruyamazawa located at the eastern part of Usoriko, and from Lake Inawashiroko (37°29′N, 140°05′E) and the Tenryu River (35°27′N, 137°50′E). The samples of the fish *T. hakonensis* were collected from Lake Usoriko, Lake Inawashiroko, and the Tenryu.

Usoriko (pH 3.4–3.8) and Inawashiroko (pH 5.0) are acidified by sulfuric and hydrochloric acid of volcanic origin, and fry of *T. hakonensis* grow to adulthood in the acid water (Satake 1980; Satake et al. 1995). The Tenryu is a typical neutral river of Japan.

Analytical methods

The pH and temperature were measured at the sites with a portable pH meter (DKK model HPH-130, Tokyo, Japan) with a glass electrode. Water samples were collected in polyethylene bottles at several locations in each lake and river. The water samples were filtered with a 0.45-μm membrane filter.

The concentrations of major and minor elements in water were determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES) (Thermo Jarrell Ash IRIS/AP, Franklin, MA, USA). Anions were measured by ion chromatography (Yokogawa IC7000, Tokyo, Japan).

Aluminum speciation was performed by high-performance liquid chromatography with postcolumn fluorescence detection, as reported elsewhere (Tsunoda et al. 1997), and the concentrations of Al^{3+}, AlL^{2+}, and $AlL^{\leqq 1+}$ species (L shows ligands combined with aluminum) were determined. Zinc and fluorine were reported to interfere with aluminum speciation by this method (Tsunoda et al. 1997). However, the effect of these elements on our analysis was negligible, because the concentrations of zinc and fluorine in water in Lake Usoriko were less than 30 ppb in our preliminary measurement.

Purified water (Organo PURIC Model-S, Tokyo, Japan) was used throughout the experiment. All bottles, flasks, beakers, and sample cups were soaked in dilute nitric acid for at least 24 h and rinsed with purified water. Gills, muscle, bone, liver, kidney, and intestine of the fish were dissected out under anesthesia. Five to seven fish from each site were analyzed. The dissected organs were weighed and dissolved in a quartz beaker using nitric acid (Cica Merck Ultrapur; Kanto Chemical, Tokyo, Japan) in a closed chamber in order to minimize any contamination. The samples were finally diluted with purified water. Aluminum concentrations were determined by a 989QZ atomic absorption spectrometer (AAS) (NJA Unicam, Cambridge, UK) with a Zeeman-effect background correction system equipped with a GF90 plus graphite furnace atomizer. The elements in the organs were analyzed using ICP-AES.

Results and discussion

Table 1 shows the pH and the major cations and anions in the water of Lake Usoriko, the acidic hot springs, and the inflowing neutral stream Maruyamazawa to show the characteristics of the water of Usoriko as a result of the mixture of strongly acidic water from the hot springs and neutral water from the catchment. Table 2 shows the pH and the major cations and anions in Lake Inawashiroko and the Tenryu River to show the large difference in water quality among these fresh waters.

The total concentration of soluble aluminum in the water was $0.51\,mg\,l^{-1}$ in Usoriko at station A, where the lake water outflows as the Shozugawa River. The total concentration of aluminum at station B, Gokurakuhama, where the water is more acidic than in the central part of the lake (Satake et al. 1999), was higher than that at station A. The total aluminum concentration differed according to site in Inawashiroko and was about $0.05\,mg\,l^{-1}$ at the center of the lake. The concentration of total aluminum in the Tenryu was very low ($0.017\,mg\,l^{-1}$). The chemical forms of aluminum in the acid water (Usoriko and Inawashiroko) are determined as the inorganic form (Al^{3+}) and the complexed form (AlL^{2+} and AlL^{1+}; L shows ligands combined with aluminum), and more than 90% of the aluminum in the water of Usoriko was measured as Al^{3+} (Table 3). However, AlL^{2+} was the dominant form in Inawashiroko.

The concentrations of major elements (Na, K, Ca, Mg, and P), aluminum, and minor elements (Fe, Cu, and Zn) in the organs of *T. hakonensis* are shown in Tables 4 and 5, respectively. The concentration of aluminum in the organs

Table 1. Water chemistry of Lake Usoriko and inflowing neutral river

Sampling site	Temp. (°C)	pH	Na	K	Ca	Mg	Fe	Al	Cl^-	SO_4^{2-}
			$mg\,l^{-1}$							
Lake water										
Sta. A	13.9	3.6	18.1	2.0	9.2	1.8	0.65	0.51	39	49
Sta. B1	14.0	3.3	23.1	2.3	9.8	1.8	0.86	0.6	54	57
Sta. B2	14.4	3.0	21.9	2.4	9.9	1.9	0.98	1.4	46	62
Sta. B3	14.9	3.2	47.4	5.2	13.8	2.9	1.57	1.1	120	80
Sta. B4	14.5	3.0	24.6	2.7	10.9	2.0	2.83	3.7	48	130
Acidic hot spring										
A	60	2.0	1803	251	169	3.7	10.4	14.3	2800	620
B	60	2.0	1714	230	161	4.1	11.6	13.9	2700	650
C	60	2.0	1494	178	158	3.2	23.7	19.5	2300	1300
Inflowing stream (Maruyamazawa)										
Sta. A	9.9	7.1	5.5	0.33	4.4	1.2	0.01	n.d.	12	2.8

Table 2. Water chemistry of Lake Inawashiroko and the Tenryu River

Sampling site	pH	Na	K	Ca	Mg	Fe	Al	Cl	SO_4^{2-}
		$mg\,l^{-1}$							
Lake Inawashiroko	5.2	5.0	0.9	8.9	2.0	0.01	0.12–0.30	11	34
Tenryu River	7.7	6.2	1.5	17.7	2.9	0.01	0.02	8.3	12

Table 3. Concentration and chemical form of soluble aluminum in Lake Usoriko, Lake Inawashiroko and the Tenryu River

Sampling site	pH	Concentration of soluble aluminum ($mg\,l^{-1}$)			
		$AlL^{\leqq 1+}$	AlL^{2+}	Al^{3+}	Total
Lake Usoriko					
Sta. A	3.6	n.d.[a]	0.04	0.47	0.51
Sta. B1 (Gokurakugahama)	3.3	n.d.	0.04	0.56	0.60
Sta. B2 (Gokurakugahama)	3.0	n.d.	0.03	1.33	1.36
Sta. B3 (Gokurakugahama)	3.2	n.d.	0.05	1.06	1.11
Sta. B4 (Gokurakugahama)	3.0	n.d.	0.03	3.66	3.69
Lake Inawashiroko					
Sta. A (center of the lake)	5.2	0.01	0.03	0.01	0.05 (0.12)[b]
Sta. B (off the coast)	5.2	0.02	0.08	0.04	0.14 (0.19)
Sta. C	5.2	0.02	0.05	n.d.	0.07 (0.18)
Sta. D	5.1	0.01	0.08	0.05	0.14 (0.20)
Tenryu River					
Sta. A (Kawakudari)	7.7	n.d.	n.d.	n.d.	n.d. (0.017)

[a] n.d.: $<0.01\,mg\,l^{-1}$

[b] Value in parentheses were determined by inductively coupled plasma atomic emission spectroscopy and graphite furnace atomic absorption spectrometry

of *T. hakonensis* collected from Usoriko was $42\,\mu g\,g^{-1}$ wet weight in gills, $4.2\,\mu g\,g^{-1}$ in muscle, $6.9\,\mu g\,g^{-1}$ in bone, $12.7\,\mu g\,g^{-1}$ in liver, $6.0\,\mu g\,g^{-1}$ in kidney, and $6.0\,\mu g\,g^{-1}$ in intestine (Table 5). The concentration of aluminum in the organs of *T. hakonensis* in Inawashiroko was roughly the same, with particular accumulation in the gills, whereas $2\,\mu g\,g^{-1}$ was found in the gills of *T. hakonensis* in the Tenryu. Thus, the concentrations of aluminum in the analyzed organs of *T. hakonensis* in acidified lakes were higher than those in the corresponding organs, especially the gills, of fish from the neutral river. This means that the gills, which are the primary organs for ion and gas exchange, consisting of a thin epithelium with a large area in contact with water, are the main target organ for aluminum toxicity in these fish. There

Table 4. Contents of major elements in the organs of *Tribolodon hakonensis*

Site and organs sampled	Na	K	Ca	Mg	P
	$mg\,g^{-1}$ (wet weight)				
Lake Usoriko (June 1997; 7 fish samples)					
Gill	1.1 ± 0.2	1.3 ± 0.1	8.6 ± 1.9	0.39 ± 0.05	6.3 ± 0.9
Muscle	0.44 ± 0.07	4.52 ± 0.03	0.61 ± 0.18	0.30 ± 0.01	2.6 ± 0.1
Bone	0.70 ± 0.18	0.73 ± 0.19	32.5 ± 5.2	0.62 ± 0.07	15.6 ± 2.3
Liver	0.93 ± 0.14	4.2 ± 0.3	0.07 ± 0.07	0.18 ± 0.02	2.5 ± 0.3
Kidney	1.5 ± 0.2	3.4 ± 0.2	0.22 ± 0.22	0.16 ± 0.01	2.5 ± 0.1
Intestine	1.3 ± 0.2	3.8 ± 0.4	0.31 ± 0.24	0.19 ± 0.02	2.7 ± 0.2
Lake Inawashiroko (May 1997; 6 fish samples)					
Gill	1.0 ± 0.2	1.1 ± 0.1	10.0 ± 2.3	0.41 ± 0.03	7.2 ± 0.9
Muscle	0.30 ± 0.07	3.7 ± 0.3	0.70 ± 0.31	0.37 ± 0.02	3.3 ± 0.1
Bone	0.97 ± 0.30	0.75 ± 0.28	31.7 ± 3.4	0.59 ± 0.04	15.6 ± 1.3
Liver	0.85 ± 0.19	2.1 ± 0.3	0.09 ± 0.06	0.25 ± 0.03	3.8 ± 0.4
Kidney	1.2 ± 0.2	1.6 ± 0.2	0.12 ± 0.06	0.17 ± 0.02	2.7 ± 0.3
Intestine	0.85 ± 0.12	2.4 ± 0.6	0.13 ± 0.07	0.24 ± 0.02	3.2 ± 0.5
Tenryu River (Dec. 1996; 5 fish samples)					
Gill	1.8 ± 0.4	1.6 ± 0.1	12.4 ± 2.4	0.45 ± 0.06	8.7 ± 1.7
Muscle	0.45 ± 0.13	3.6 ± 0.4	0.75 ± 0.20	0.40 ± 0.04	3.3 ± 0.2
Bone	2.2 ± 0.6	0.70 ± 0.15	58.7 ± 9.6	0.95 ± 0.12	28.8 ± 4.5
Liver	0.87 ± 0.02	2.9 ± 0.6	0.07 ± 0.01	0.20 ± 0.03	3.2 ± 0.1
Kidney	1.4 ± 0.1	3.1 ± 0.3	0.12 ± 0.04	0.21 ± 0.02	3.5 ± 0.3
Intestine	1.0 ± 0.2	2.3 ± 0.4	0.13 ± 0.05	0.25 ± 0.05	3.0 ± 0.5

Table 5. Contents of aluminum, iron, copper, and zinc in the organs of *Tribolodon hakonensis*

Site and organs sampled	Al	Fe	Cu	Zn
	$\mu g\,g^{-1}$ (wet weight)			
Lake Usoriko (June 1997; 7 fish samples)				
Gill	42 ± 23	42 ± 16	1.2 ± 0.2	25.3 ± 2.9
Muscle	4.2 ± 2.6	5.5 ± 1.6	0.67 ± 0.07	5.0 ± 0.6
Bone	6.9 ± 2.7	7.0 ± 3.6	0.49 ± 0.03	31.9 ± 5.6
Liver	12.7 ± 5.8	412 ± 180	35 ± 30	27.9 ± 7.0
Kidney	6.0 ± 1.5	110 ± 28	2.8 ± 0.4	33.5 ± 5.2
Intestine	6.0 ± 1.2	128 ± 69	2.5 ± 0.5	26.1 ± 3.9
Lake Inawashiroko (May 1997; 6 fish samples)				
Gill	37 ± 22	46 ± 20	0.73 ± 0.27	26.6 ± 3.3
Muscle	5.0 ± 3.0	5.2 ± 0.9	0.52 ± 0.30	7.0 ± 1.8
Bone	19.5 ± 2.2	4.6 ± 1.4	0.20 ± 0.02	37.4 ± 0.7
Liver	18.3 ± 8.7	277 ± 65	22 ± 14	32.1 ± 2.7
Kidney	7.2 ± 4.0	102 ± 30	2.3 ± 0.6	64.1 ± 20.7
Intestine	6.3 ± 3.3	77 ± 41	1.8 ± 0.7	30.7 ± 4.6
Tenryu River (Dec. 1996; 5 fish samples)				
Gill	1.6 ± 0.7	29.6 ± 4.5	0.66 ± 0.12	22.8 ± 2.3
Muscle	1.1 ± 0.2	8.7 ± 6.2	0.70 ± 0.37	9.0 ± 1.8
Bone	2.5 ± 1.1	2.9 ± 0.6	0.34 ± 0.09	42.1 ± 3.5
Liver	5.8 ± 3.1	167 ± 28	14 ± 13	22.9 ± 3.4
Kidney	3.0 ± 1.2	118 ± 33	1.6 ± 0.3	30.6 ± 4.6
Intestine	0.5 ± 1.0	23 ± 8	1.9 ± 0.5	23.5 ± 3.4

have been many reports on the effect of a high concentration of aluminum in acidic waters on fish, and most have focused on damage and aluminum accumulation in the gills (Karlsson-Norrgren et al. 1986a,b; Youson and Neville 1987; McCahon et al. 1989; Vuorinen et al. 1990; Eeckhaoudt et al. 1996). However, the accumulation of aluminum in organs has not been well studied, and it is unclear whether aluminum is taken into the bloodstream and distributed to the body. This may be partly because of analytical problems with the measurement of aluminum in biological tissues. Since aluminum levels are usually low in biological tissues, sensitive analytical methods are required. In addition, aluminum contamination can easily invalidate the analytical results (Tölg 1993). Karlsson-Norrgren et al. (1986a), however, determined the concentrations of aluminum in various organs (gills, kidney, liver, intestine, muscle, and bone) of brown trout in acidified farms. They reported that exposure to aluminum was reflected by relatively high

Table 6. Bioaccumulation factors of aluminum in various fish organs (ml g^{-1})

Organs	Lake Usoriko	Lake Inawashiroko	Tenryu River
Gill	70	740	94
Muscle	7	100	65
Bone	12	390	150
Liver	21	370	340
Kidney	10	140	180
Intestine	10	130	29

aluminum concentrations in many of the organs, especially the gills, and the aluminum concentration was high in the kidney, among the internal organs. In that study, gill lesions were observed, and the reported aluminum concentrations were 20 and 90 μg g^{-1} wet weight in the gills and 1–10 μg g^{-1} wet weight in the other organs.

The aluminum concentrations we obtained were of a similar order as those obtained by Karlsson-Norrgren et al., but the pattern of aluminum accumulation differed. We found that the hepatic levels were higher than the renal levels, even in the control fish living in neutral water. The adult *T. hakonensi*s we examined are considered to live a healthy life in naturally acidified lakes. Even though the aluminum concentrations in the gills were very high, no disorder was detected. *T. hakonensis* in Usoriko has a different type of chloride cell in the gills, which enables it to live in the strongly acidic water (Mashiko et al. 1973), because *T. hakonensis* living in neutral water does not show acid tolerance. The regulation of protons, which may be controlled by chloride cells, and of aluminum will have to be considered.

The bioaccumulation factors (BAFs) of aluminum in various fish organs, calculated by dividing the wet tissue concentration by the water concentration, are shown in Table 6. The BAFs of organs of fish in Usoriko were smaller than those of the corresponding organs of fish in Inawashiroko and the Tenryu River. Examination of the aluminum species in water showed that more than 90% of the aluminum in water from Usoriko was Al^{3+}, whereas AlL^{2+} was dominant in the water from Inawashiroko. Previous studies have indicated that the low-molecular-weight inorganic form of aluminum (Al^{3+}), often referred to as inorganic monomeric aluminum, appears to be the most toxic species (Driscoll et al. 1980). Our results suggested that there is a difference between the uptake of toxic Al^{3+} and the complexed form (AlL^{2+} or other complexed forms, which are less toxic). The aluminum concentrations in the organs of *T. hakonensis* in Usoriko and Inawashiroko were roughly the same except for that in bone, despite the difference in water quality. This means that the aluminum concentration in the body of the fish does not directly reflect the concentration in the water. This in turn suggests that accumulation of aluminum in the fish is somehow controlled by chemical and biological factors in these acidified lakes.

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