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Influence of Complexans (NTA, EDTA) on the Toxicity of Aluminum Chloride and Sulfate to Fish at High Concentrations

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There are several reports on the acute toxicity of Aluminum salts to aquatic animals. Specific studies on the toxicity of aluminum chlorides to the copepod (*Nitcra spenipes*) were examined by BERGTSSON (1978). The acute toxicity of aluminum chloride on the survival of three species of polychaetes was determined by STEPHEN et al. (1979). EBELING (1928) reported that 5.0 ppm aluminum sulfate solutions caused a trout to turn over in 5 min, and THOMAS (1915) stated that 7.0 ppm aluminum sulfate killed mummichogs in 5 days. However, information on the effects of complexing agents on the metal-toxicity to the fish is insufficient. In the previous paper (MURAMOTO 1980a,b; 1981), the effects of complexans (EDTA, NTA and DTPA) on the metal-toxicity (Cd, Cu, Zn and Pb) at lethal levels and of EDTA on the removing cadmium from the Cd-contaminated fish, and also the influence of complexans (EDTA and DTPA) on the toxicity of cadmium to fish at chronic levels were reported.

The present experiment studied the effect of complexans (NTA,EDTA) on the toxicity of lethal levels of aluminum chloride and aluminum sulfate to fish.

MATERIALS AND METHODS

Experimental conditions

Each group of ten carp (*Cyprinus carpio* L.) 7.5-8.5 cm in length and weighing 10.5-12.5g, was kept in a 60-L glass container at 14.5-16.5°C throughout the experiment. These were altogether 21 such groups: six groups each containing aluminum chloride ($AlCl_3.6H_2O$) or sulfate ($Al_2(SO_4)_3.18H_2O$) alone at concentrations of 2.0, 4.0 and 8.0 ppm; twelve groups each containing 2.0, 4.0 or 8.0 ppm of aluminum chloride or sulfate together with an equimolar concentration of one complexan, either the trisodium salt of nitrilotriacetic acid (NTA) or the tetrasodium salt of ethylenediaminetetraacetic acid (EDTA), 3 times those of the metals; two containing complexan alone; and one control environments.

Analysis

All ten fish from each test group were dissected into three parts: viscera, gills, and other parts. Each of these parts was incinerated at 450°C for 24-h in an electric muffle furnace. The ash sample was

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dissolved in the mixed acid HNO_3-$HClO_4$(2:1), and made up to a fixed volume by addition of 0.1N-HCl. Al was determined using an atomic absorption spectrophotometer. The fuels used were acetylene-nitrous oxide.

RESULTS AND DISCUSSIONS

Mortality of fish

The mortality rate(%) of fish after each 24-h period and the pH of breeding water are shown in Table 1. Mortality increased moderately with an increase in the concentration of metal in water. No fish deaths occurred in environmnets containing 2.0 ppm of aluminum chloride and sulfate alone. Mortality in the 4.0 ppm and 8.0 ppm Al groups increased with time, reaching 30 and 50 % in the aluminum chloride group and 10 and 30 % respectively in the aluminum sulfate group within 48-h. In contrast, addition of complexans decreased mortality by 10 % in the 4.0 ppm aluminum chloride-plus-EDTA group, and 20 % in the 8.0 ppm aluminum sulfate-plus-NTA group within 48-h. The Al accumulation in the gills was 1.5-8.9 times greater than those in the viscera for the studies involving aluminum chloride and 1.3-9.6 times greater for those studies involving aluminum sulfate. Aluminum chloride was equivalent in toxicity to cadmium (MURAMOTO 1980b) and more toxic to carp than aluminum sulfate.

Table 1. Mortality (%) of fish after 24 and 48-h and initial pH of breeding water

Al compounds	$AlCl_3.6H_2O$			$Al_2(SO_4)_3.18H_2O$		
Experimental group (ppm)	pH	24-h	48-h	pH	24-h	48-h
Al 2.0	6.6	0	0	6.7	0	0
Al 2.0 + EDTA 100	6.8	0	0	6.7	0	0
Al 2.0 + NTA 61	6.8	0	0	6.8		
Al 4.0	6.5	10	20	6.6	0	10
Al 4.0 + EDTA 201	6.6	0	10	6.7	0	0
Al 4.0 + NTA 122	6.7	0	0	6.7	0	0
Al 8.0	6.3	30	20	6.4	20	10
Al 8.0 + EDTA 402	6.5	0	0	6.6	0	0
Al 8.0 + NTA 244	6.6	0	10	6.6	10	0
EDTA 402	7.5	0	0			
NTA 244	7.4	0	0			
Control	6.9	0	0			

Comparison of the Al concentrations in the water and in the 48-h surviving fish

The Al content (μg/g in ash) in the viscera, gills and other parts of the fish for 48-h with or without complexans are shown in Fig. 1. A positive correlation was observed between the concentration of Al in the water and that in the each parts of the fish. The concentration of Al increased in the following order : gills>viscera>other parts. The concentrations was remarkably high in the gills.

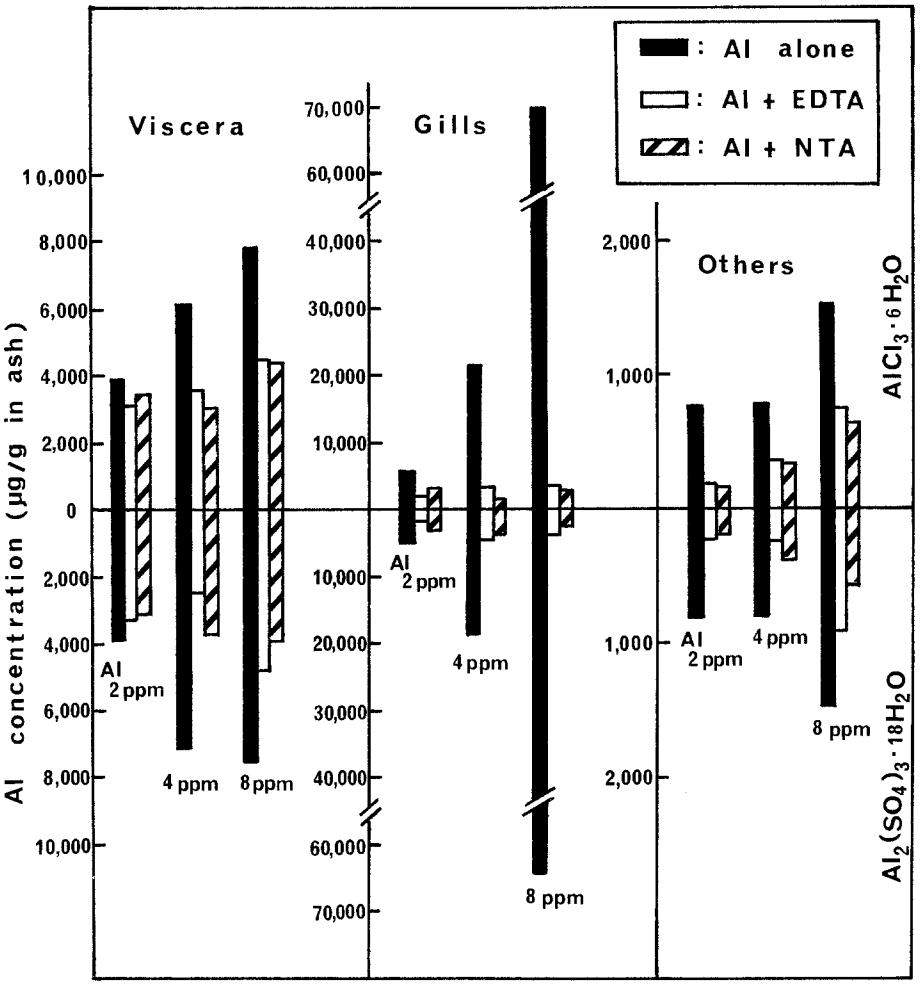

Fig. 1 Al concentrations (μg/g in ash) in each parts of the 48-h survived fish

The Al accumulations in the aluminum chloride group compared favorably to with those in the aluminum sulfate group : 0.84-1.2 times for viscera, 1.1-1.2 times for gills, and 0.95-1.0 times for other parts, respectively. The Al concentrations in the gills of the fish exposed to aluminum chloride were slightly higher than the concentration in the fish exposed to aluminum sulfate. On the other hand, the ratio (%) of decreases of Al concentrations in each parts of the fish in metal-plus-complexan group compared with those in the metal-alone group was as follows : as to the aluminum chloride group treated with either EDTA or NTA, the decreases-ratio was 23-42%, 12-43% for viscera, 64-95%, 47-95% for gills, and 53-74%, 57-76% for other parts, respectively, and as to the aluminum sulfate group, those was 15-37%, 19-46% for viscera, 62-94%, 39-96% for gills, and 39-72%, 54-80% for other parts. Also, the Al concentrations of the fish in EDTA or NTA alone group were all less than 1.0 μg/g Al in ash in each parts of the fish.

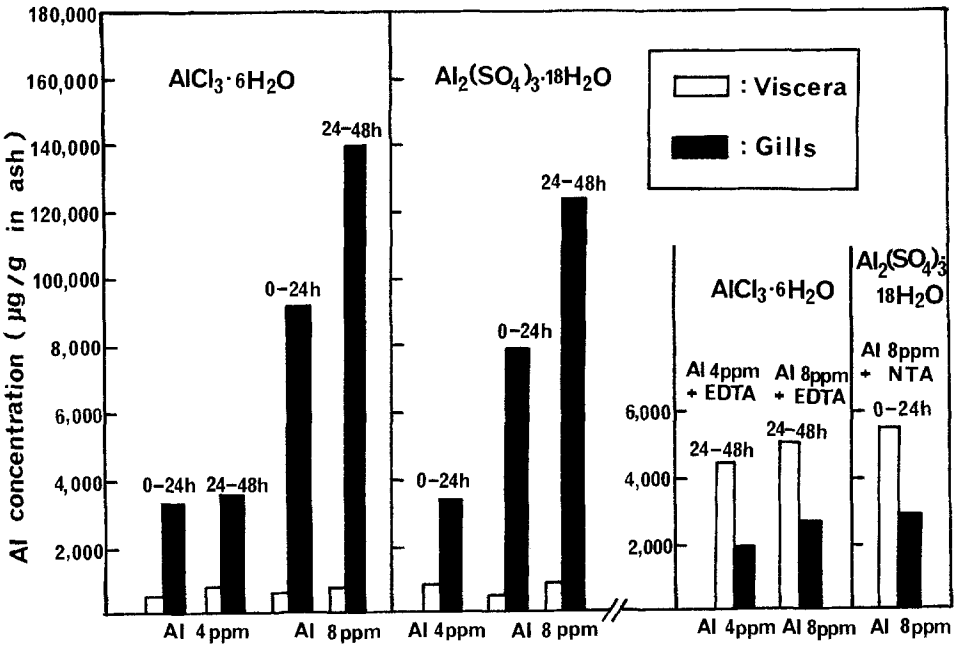

Fig. 2 Comparison of the Al concentrations (μg/g in ash) in viscera and gills between the dead and the 48-h survived fish

Comparison of tissue metal concentration in ash between the dead and the 48-h survived fish

The tissue metal concentrations between the dead and the 48-h survivors are shown in Fig. 2. In the aluminum alone group, the Al concentrations (μg/g in ash) of the dead fish compared with that in the 48-h survivors were : 0.82-1.6 times and 0.88-1.5 times in the viscera for either aluminum chloride and aluminum sulfate, and 1.3-2.0 times, 1.2-1.9 times in the gills for aluminum chloride and aluminum sulfate, respectively. This suggested that the dead fish was caused by adsorption of aluminum metal on the gills tissues. In contrast, the inhibitors of metal accumulation by the presence of complexans was remarkably indicated in the gills, however, no significantly decreases of the Al accumulation was occurred in the metal-plus-complexan group according to the increases of the Al accumulation in the viscera by addition of the complexans either EDTA or NTA. Whereas, the Al concentrations of the dead fish in the metal-plus-complexans group compared with those in the metal alone group was as follows : 0.65-0.73 times in the viscera, 0.04-0.09 times in the gills for aluminum chloride-plus-EDTA, and 0.81 times in the viscera, 0.05 times in the gills for aluminum sulfate-plus-NTA group, respectively. This indicated inhibitions of the metal accumulation and the metal toxicity by adding the complexans. From these results, it is suppose that the aluminum can pass through the gills as metal-complexes by the presence of complexans.

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REFERENCES

BERGTSSON,G.E. : Marine Poll. Bull. 9, 238 (1978).

EBELING, G. : Zeits. Fisherei, 26, 49 (1928).

MURAMOTO,S.: Bull. Environm. Contam. Toxicol. 25, 828 (1980a).

MURAMOTO,S.: Bull. Environm. Contam. Toxicol. 25, 941 (1980b).

MURAMOTO,S.: Bull. Environm. Contam. Toxicol. 27, (1981), in press.

STEPHEN,M.P.&J.R.DONALD: Bull. Environm. Contam. Toxicol.23,698(1979).