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Toxicity Assessment of Contaminated Sediments after Dredging

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Contaminated sediments are considered as high-priority risks to environments and human health. Remediation by dredging is a commonly used measure to remove these hazards (Murphy et al, 1999). However, dredging of contaminated sites may result in resuspension and release of toxic pollutants, which can potentially cause an ecotoxicological risk for an aquatic system (Hyötyläinen and Oikari, 1999).

In the past decades, Taihu Lake of China has been seriously polluted by industrial and rural effluents, and agricultural runoff, rich in phosphorus (Zuo et al., 2003), organic contaminants and heavy metals (Zou et al., 1996; Wang et al., 2003). This has resulted in severe degradation of water quality and frequent break out of water bloom in Taihu Lake. At present, contaminated sediments of Taihu Lake became a major concern to people after control of external sources. Therefore, as a remedial tool, in 2003, sediment dredging was carried out in Wulihu Lake, a lagoon of Taihu Lake, China, to speed up the recovering of the Lake. This paper, as a part of a wider research program on the monitoring of the Lake recovery, will assess the toxicity potential of dredged Wulihu Lake sediments by using the microbiological test systems.

MATERIALS AND METHODS

Sediments were collected monthly from Nov/03 (one month before dredging) to Dec/04 (a year after dredging) with a 0.05 m^2 polyethylene grab at Wulihu Lake, a lagoon at the north top of Taihu Lake, China. The collected samples were transported to a laboratory at appreciably 4 ℃ with a sealed polyethylene bag. After removing plants liters, large rocks and shells in the samples, all samples were freeze-dried with ALPHA 1-2LD (Christ, Germany) and homogenized for trace element analysis and toxicity test all together at the end of the experiments.

Aliquots of the sediment samples were digested with HNO_3 and HCl at 95 ℃ according to the US EPA Method 3050B (US EPA, 1999). The analysis of trace metals was performed using inductively coupled plasma atomic emission spectrometry (ICP/AES, Perkin-Elme CO. U.S.A). Analytical reagent blanks were prepared with each batch of the digestion set and then analyzed for the same elements as the samples.

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Elutriate preparation: 10.00 g of sediment sample was shaken vigorously with 10 mL of 3 % NaCl solution for 1h at room temperature. Then the sediment was settled overnight at 4 ℃ and the supernatant was filtered (0.45 μm, Wattman GF/C) under vacuum to eliminate colloidal particles or suspended solids in the solution, which can cause an optical interference during the luminescence measurement.

The potential toxicity of sediment elutriates were tested according to Chinese National Standard (GB/T15441-1995, China). The toxic effects were monitored as a percent decrease of the light emission of *Photobacterium phosphareum* T3 after 15 min of incubation. All assays were conducted with the DXY-2 Biological Toxicity Analyzer (Institute of Soil Science, Chinese Academy of Sciences). The nutrient broth was culture medium of test organism. Bacterial suspensions were equilibrated to 15 ℃ after rehydration. The pH value of the sample was adjusted to 7.0 ± 0.2. Each dilution step, each concentration of environmental samples and the controls were tested in 4 duplicates. The end point was the 15 min EC_{50}, expressed as dry weight of sediments.

Bacteria contact assays for sediment samples were performed according to Heise and Ahlf (2005). All assays were performed with a 96-well microplate meter (GENios Plus, TECAN Inc, Germany). *Photobacterium phosphareum* T3 was the test organism. In brief, a 10.00 g sample was weighted into a beaker containing 10 mL 3% NaCl and diluted to prepare the highest test concentration of 50 % dry weight of sediments. A dilution series was set up in which this suspension was diluted 1:2 with a quartz sand/saline suspension of the same dry weight. Six concentrations (25, 12.5, 6.25, 3.13, 1.6, 0.8 % dry weight of sediments) and two controls (bacterial + quartz sand, sediment suspension only) comprised the experimental configuration. The bacterial suspension (OD_{600} = 0.1), a dilution of an overnight culture (28 ℃) with broth medium, was used as inoculums, which was exposed in quadruple parallels to each concentration step. 100 μL sediment suspensions and 100 μL bacterial suspensions were added to each well of the test plates, which were incubated on an overhead shaker at 28 ℃ for 2 h. And afterward 40 μl of a resazurin solution (10 mg /100 mL distilled water) were added to each well, the plates were incubated and shaken again. Endpoint of the test was the resorufine production in the test wells compared to the resorufine production in the control test. Fluorescence of resorufine was measured at the excitation/emission wavelengths 560/590 nm. Calibration curves for each sediment concentration were calculated. In the calibration plates, the different sediment suspensions were supplemented with 100 μL of sterile culture medium and mixed with solutions of different resorufine/resazurine- ratios (20/80, 40/60, 50/50, 60/40, v/v).

According to Blaise and Ménard (1998), Toxic effects on algae were determined by measuring the esterase enzymatic activity of *Selenastrum capriconutum* (UTEX 1648), supplied by the Freshwater Algae Collection of Institute of Hydrobiology, Chinese Academy of Sciences. The bioassay was performed in 20 × 150 mm glass tubes containing a starting concentration of 10 % dry weight of sediment sample to which was added a unicellular inoculum of the algae (10^6

cells/mL). Five concentration steps of the test sediment (10, 5, 2.5, 1.25, 0.625 % of dry weight of sediment), and three controls (algae only, test sediment only and algae + acid-washed quartz sand), each run in triplicate tubes, comprised the experimental configuration. All tubes were capped loosely with silica gel plug and placed in a vertical rotator (20-25 rpm, 22 ± 0.2 ℃, 24 h). After exposure, tubes were settled for 5 min, and then 1 mL supernatant was withdrawn from each tube and dispensed into an acid-washed plastic tube, used especially for flow cytometry. 30 μL of a 1000 mM fluorescein diacetate (Sigma, St. Louis, MO, dissolved in acetone) was micropipetted into each tube. After 15 min incubation in darkness, the fluorescence of the algal cell was assessed via a FACSVantage SE flow cytometer (Becton Dickinson, USA). The end point was a 24 h EC_{50}, expressed as percent weight of wet sediment (w/v). A nontoxic response with this assay corresponded to an EC_{50} > 10 % (w/v) of whole sediment.

In this paper, EC_{50} values and associated 95 % confidence intervals were calculated using Probit analysis with statistical software SPSS12.0 (SPSS Inc., USA). The significance between the EC_{50} values of sediment samples were determined using the method described by Sprague and Fogels (1977).

RESULTS AND DISCUSSION

To assess the ecotoxicological sense of trace element concentrations in the pre- and the post-dredging sediments, two sets of sediment quality guidelines (SQGs) developed for freshwater sediments (Simth et al, 1996; MacDonald et al, 2000) were applied in this study: (a) the threshold effect level (TEL)/probable effect level (PEL) values and (b) the threshold effect concentrations (TEC)/ probable effect concentrations (Table 1). Low range values (i.e., TELs or TECs) are concentrations below which adverse effects upon sediment dwelling fauna would infrequent be expected. In contrast, the PELs and PECs represent chemical concentrations above which adverse effects are likely to occur (MacDonald et al, 2000). Considering the results, given in Table 1, sediments collected before dredging were toxic as concentrations of Cr and Ni exceeded their PEL values. After dredging, they were less toxic due to no significant exceedance of PEL or PEC value for all analyzed trace elements; that is, sediments with respect to Cu, Zn and Pb were grouped lower TEL; with respect to Cr and Ni they were grouped into TEL-PEL/TEC-PEC, although they were still grouped above PEL at Jun 2004 and Sep 2004 with respect to Ni, which might be attributed to sedimentation of resuspended contaminated sediments after dredging or external loading of industrial effluents. These results indicated that adverse effects would still be likely to occur with dredged sediments.

The incidence of toxicity was determined among all sediment samples collected at pre- and post-dredging by using luminescent bacteria *Photobacterium phosphareum* T3 and freshwater chlorophyte *Selenastrum capriconutum* UTEX 1648 as test organisms. The result of bioassays performed with sediment elutriates and bulk sediments were summarized in Table 2.

Table 1. Trace elements contents in sediments and TEL, PEL, ERL, ERM guideline values (X ±SD; μg/g dry weight)

Sediments	Copper	Zinc	Chromium	Lead	Nickel
Nov/03	57.0±4.1	175.3±16.5	93.6±2.1	82.4±4.2	76.8±8.5
Dec/03	59.8±5.6	140.7±14.3	94.9±4.9	76.6±7.4	73.6±3.6
Feb/04	37.6±5.5	93.2±6.2	77.1±9.2	69.5±9.7	33.7±3.8
Mar/04	28. 7±2.7	67.8±12.1	69.2±7.8	62.2±6.4	31.4±5.6
Jun/04	21.8±2.2	53.4±12.0	70.7±9.8	58.9±3.1	36.3±3.5
Sep/04	21.5±1.4	48.4±9.6	74.2±2.9	56.8±9.8	36.6±1.6
Nov/04	19.5±2.2	44.9±7.9	76.8±1.2	51.7±6.4	35.7±2.8
Dec/04	19.7±1.9	45.5±7.8	63.9±3.9	61.9±6.8	29.3±4.5
TEL [a]	37.5	123	37.3	35	18
PEL [a]	197	315	90	91.3	36
TEC[b]	31.6	121	43.4	35.8	22.7
PEC[b]	149	459	111	128	48.6

Nov/03 and Dec/03 were before dredging; Feb/04~Dec/04 were after dredging; [a]Simth et al., 1996; [b]MacDonal et al, 2000

As shown in Table 2, based on EC_{50} determinations, a gradient of increasing toxicity response were observed in going from Nov/03 (EC_{50} = 5.82 %), Dec/03 (EC_{50} = 4.45 %) to Feb/04 (EC_{50} = 2.85 %). This result might indicate that sediment dredging could induce an increase of sediments bacterial toxicity for a short time, which could be attributed to increased bioavailability of sediment trace

Table 2. Bioassays acute toxicity of sediments elutriates and the whole sediments (% dry weight of sediment, w/v)

Sediments	Elutriate tests		Bacteria contact assay		Algal solid phase assay	
	EC_{50}	95% confidence interval	EC_{50}	95% confidence interval	EC_{50}	95% confidence interval
Nov/03	5.82	4.69-8.69	5.10	3.50-20.8	4.51	2.19-6.93
Dec/03	4.45	1.65-10.66	5.58	4.15-9.61	5.42	2.33-10.64
Feb/04	2.85	0.069-9.31	3.89	2.62-7.88	3.39	2.73-4.38
Mar/04	18.64*	11.03-34.98	4.15	3.05-5.88	2.31*	1.73-3.17
Jun/04	9.82*	5.80-17.01	3.24	1.95-7.49	3.06	2.03-5.41
Sep/04	22.57*	16.15-35.02	4.36	3.22-6.15	4.50	3.58-5.96
Nov/04	16.05*	9.67-35.75	4.63	2.26-134.6	ND	ND
Dec/04	8.62	1.30-37.06	4.68	3.69-6.79	1.41*	0.30-2.80

Nov/03 and Dec/03 were before dredging; Feb/04~Dec/04 were after dredging; *, significance between the EC_{50} values of sediment sample in pre- and post-dredging; ND: not determined

metals with sediment dredging (Wirth et al, 1996). However, comparison with the EC_{50} values of the pre-dredging sediments (Nov/03 and Dec/03), significant decreasing toxicity responses were observed after one month of sediment

dredging, while the majority of the sediment samples were also considered toxic according to Onorati et al. (1998), which might be indicate that bacterial toxicity of sediment elutriates was lower after sediment dredging.

A bacterial contact assay was conducted to assess potential toxicity of the whole-sediment samples collected at pre- and post-dredging. Table 2 showed EC_{50} values for all sediment samples after 2 h of incubation of *Photobacterium phosphareum* T3. Comparison of EC_{50} values between sediments showed that sediments collected before dredging usually yielded greater EC_{50} values than those collected after dredging, indicating that sediments were more toxic after dredging. The explanation for this result might be increased bioavailability of contaminants with sediment dredging disturbance (Wirth et al, 1996). But this result was inconsistent with that of the sediment elutriates tests. In the other hand, the EC_{50} values of bacterial contact assay were distinctly lower than those of elutriates tests after dredging, although the test organism of both assays were the same one. This difference might be caused by the contaminants those were not extracted by water but still available to test organism. It might indicate that the bacterial contact assay was more suited to assess the toxic effects of bulk sediment (Guzzella, 1998).

The algal solid-phase assay (ASPA) is a micro-algal test designed to assess the toxic potential of freshwater sediment (Blaise and Me'nard, 1998). The principle of this test is based on esterase enzymatic activity (and its inhibition), which rapidly measures the physiological status of cells after exposure for 24 h to solid media samples. The application of esterase end point to determine the toxicity of industrial effluents and inorganic contaminants to micro-algae has demonstrated the usefulness of this biomarker for ecotoxicological investigations (van Beelen, 2003; Adams and Stauber, 2004). In this study, the ASPA was used to determine the potential toxicity variation of sediment of Wulihu Lake after dredging. As shown in Table 2, all sediment samples collected before and after dredging consistently yielded EC_{50}< 10% dry weight of sediment, suggesting they were all toxic. And based on EC_{50} endpoints, a general gradient of increasing toxicity responses was observed in going from Nov/03 (EC_{50}=4.51%) to Decr/04 (EC_{50}=1.41%), which might indicate that the toxicity of the sediments enhanced after dredging. The explanation for this result was the same to bacterial contact assay. This result was some similar to that of bacterial contact assay, but inconsistent to those of elutriates tests.

Bioanalytical comparison of results should be indicated that, in contrast with other microscale tests, the elutriate test displayed less toxicity toward most sediment samples collected after dredging. The difference of toxicity responses between these bioassays could be understood that the elutriate test was unsatisfactory in predicting bulk sediment toxicity, as it could not mimic the in situ exposure to aquatic organism (Guzzella, 1998). Although the bioanalytical comparison cannot elucidate the divergence in results, the qualitative differences in agreement between the elutriate test and other microscale tests may be a result of bioassays sensitivity, time-related adaptation, changes in bioavailability related to

contaminants and their forms. However, it was important that all toxicity tests confirmed the endobenthic toxicity of the sediments in Wulihu Lake after dredging.

Correlations analysis between toxicity tests and chemical data were unable to explain the toxicity responses in the bacterial contact assay or the algal tests. This result confirmed that microbiological test systems were more efficient methods for rapid screening and continuous monitoring of toxicity of contaminated sediments (Blaise et al., 2000; van Beelen, 2003). Others, the algal responses in this study may be correlated to the organic pollutants in sediment of Taihu Lake (Zou et al., 1996; Wang et al., 2003), as *S. capricornutum* was generally sensitive to a wide array of (in)organic chemicals (Sander et al., 1985). Thirdly, the ecotoxicological significance of toxic metals inWulihu Lake sediment may be determined by their physicochemical forms in sediment rather than by their total concentrations (Amiard, 1992). In this paper, the poor links between the toxicity and toxic metals also confirmed that it was unsatisfactory to evaluate the effect of sediment dredging based on the chemical data of sediments (Hyötyläinen and Oikari, 1999).

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