Ecotoxicology and Environmental Safety 122 (2015) 178–185

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

journal homepage: www.elsevier.com/locate/ecoenv

Arsenic pollution and its treatment in Yangzonghai lake in China: In situ remediation

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ARTICLE INFO

Article history:
Received 22 April 2015
Received in revised form
25 July 2015
Accepted 27 July 2015
Available online 1 August 2015

Keywords:
Arsenic pollution
Coagulation method
$FeCl_3$
Yangzonghai Lake

ABSTRACT

In this study, the effect of direct atomization and spraying a ferric chloride ($FeCl_3$) solution to decrease the arsenic concentration and its pollution in Yangzonghai Lake, China, was investigated. Ten ships were used for spraying 6–8 t of $FeCl_3$ in the lake every day since October 2009. After spraying, the average concentration of arsenic in Yangzonghai Lake, which has an area of 31 km^2, an average depth of 20 m, and a water storage capacity of 604 million m^3, started to decrease from 0.117 mg L^{-1}. On 20 September 2010, the lowest arsenic level of 0.021 mg L^{-1} was attained, with an arsenic removal rate as high as 82.0%. However, the source of pollution was not eliminated, and local rainfall mainly occurred in September; hence, arsenic concentration from October to December increased to 0.078 mg L^{-1}. At the beginning of 2011, the As concentration decreased and remained at 0.025–0.028 mg L^{-1} from May to September. During the 2 years of $FeCl_3$ treatment, the water quality improved from V Class to II–III Class of the Chinese standards, which remained consistent for 12 months. The total cost for this in situ water treatment was 29 million RMB, which was less than a hundredth of the expected expenditure of 4–7 billion RMB. The treatment method achieved goals such as high arsenic removal rate, easy operation, low cost, and ecological security. In this study, the changing patterns of the concentration of arsenic in Yangzonghai Lake from June 2008 to December 2014 were analyzed, and the following problems were discussed: the stability of iron-arsenic precipitates in the lake, the concentrations of ferric and chloride ions in the lake, the pH of the lake during treatment, the stability of iron-arsenic precipitates in the lakebed sediments, and the variation of phytoplankton species in the lake.

1. Introduction

In June 2008, Yangzonghai Lake, a large plateau lake, was polluted owing to the illegal discharge of arsenic by chemical enterprises in the Yunnan Province, China. As a result, the concentration of arsenic in lake reached 0.128 mg L^{-1} in September. The water quality decreased from Class I to V of the Chinese standard, making it impossible to use the lake water for drinking, fisheries, and agricultural irrigation. Owing to these circumstances, the Yunnan Government held a press conference on September 19 with greater than 60 media professionals, gaining attention and sharp criticism both at home and abroad. Later, three heads of the chemical enterprise were sued and sentenced, and 26 government officials were called to account and punished. After the incident of arsenic pollution, several Chinese research institutes and universities have started investigation (Wang et al., 2010; Qi et al., 2010; Zhang et al., 2010; Bi et al., 2014).

Yangzonghai Lake spans an area of 31 km^2, with an average depth of 20 m, and its water storage is 604 million m^3. There are no practical techniques or referential experiences for treating the arsenic pollution of such a huge water body anywhere in the world. On 10 October 2008, the Yunnan Province conducted a public bidding to the whole world through the Science and Technology Project of “Yangzong Lake Pollution Abatement, Arsenic Removing and Water Quality Restoration in Yunnan, China” and stipulated that the unit that wins the bidding should first attempt to decrease the concentration of arsenic in the water to 0.050 mg L^{-1}.

The biggest difficulty in treating the arsenic pollution in Yangzonghai Lake is attributed to the fact that the capacity of this water body is too large and that the method adopted must be highly efficient and cost-effective for removing arsenic as well as

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http://dx.doi.org/10.1016/j.ecoenv.2015.07.032
0147-6513/

should be easy to operate and exhibit ecological safety. Previously reported methods such as ion exchange (Vagliasindi and Benjami, 1998), different modified coagulation methods (Chwirka et al., 2004; Song et al., 2006; Andrianisa et al., 2008; Wan et al., 2011a, 2011b; Bordoloi et al., 2013), adsorption–colloid–flotation (Peng and Di, 1994), photocatalysis (Bissen et al., 2001; Dutta et al., 2005), various new material adsorption methods (Lenoble et al., 2005; Awual et al., 2012), and nanomaterial adsorption (Xu and Meng, 2011) could not be employed. Hence, in September 2009, only three alternatives were available. First is laterite adsorption (Cai et al., 2009). According to the calculation of a scale-up experiment, 250,000 t of specific laterite (iron content $>8\%$) is required, and the diameter of the soil particles needs to be ground to 0.04 mm. Then, it needs to be size-mixed and sprayed. However, this method does not exhibit a high adsorption rate, and the project quantity is huge. Second is ionic sieve adsorption (Yuan, 2009). In this method, the absorbent is wrapped into small bags, and then the bags are hung on a hull bottom, and the arsenic is adsorbed from the water through the itinerant floating of ships. Last is coagulation using $FeCl_3$, in which arsenic is in situ sealed up in the lakebed. This method uses amorphous $Fe(OH)_3$ produced by the hydrolysis of $FeCl_3$ to adsorb arsenic. Recently, the results of the scale-up experiment employing this method in a laboratory, and an engineering test of the on-spot water bodies of 10,000 m^3 and 250,000 m^3 have been reported (Chen et al., 2015). As against traditional $FeCl_3$ coagulation, this method requires neither the oxidation of As(III) to As(V) nor the adjustment of the pH of the lake water. A series of experiments on 50 L of lake water were conducted in the laboratory, and the arsenic removal rate was observed to reach up to 96.7% with the pH value remaining unchanged. The quantity of $FeCl_3$ reagent could be reduced to 1.6–3.2 mg L^{-1}. Although the quantity of reagent is very low, 1000–2000 t of $FeCl_3$ is still needed according to the calculation of a water body of 6042 million m^3. As the solubility product of $Fe(OH)_3$ is 4×10^{-38}, the concentration of Fe^{3+} in the water is lower than the detection limit (<0.010 mg L^{-1}; Tian, 2010). Irrespective of the amount of $FeCl_3$ added to the water, only the concentration of Cl^- increases, which does not affect ecology. The project operation of the method described herein is extremely simple; meanwhile, it satisfies demands such as low cost, low energy, high arsenic removal rate, as well as being environmentally friendly.

In October 2009, the project team in this paper used 10 spraying boats for carpet spraying two times per day. The average concentration of arsenic in Yangzonghai Lake has been continuously decreasing since November 2009. In September 2010, the minimum arsenic concentration of 0.021 mg L^{-1} was attained. The rainwater in September and October carries arsenic into the lake through the spring mouth on the lakeshore. These arsenic pollutants mostly originate from the underground soil, underground karst caves, and limestone lava crannies under Jin-Ye Chemical Plant (JYCP). As a result, the arsenic concentration increases and rebounds in November and December. However, after January 2011, the concentration continuously decreased again and remained steady at 0.025–0.028 mg L^{-1} between the end of April and the middle of September. After this period, the arsenic concentration increased and rebounded again from October to December because of rainwater. Spraying stopped in November 2011. The environmental protection departments in Kunming City thought that they needed a period of time to observe and evaluate the treatment scheme for immobilizing arsenic in its home position. Hence, $FeCl_3$ spraying is not continued.

A close relationship exists between lake treatment and whether arsenic pollution sources have been cut off, as well as the ways in which pollutants flow into the lake, hydrogeological conditions of the nearby areas, rainfall distribution during treatment, and current dynamic strength of the water body. In this study, the effects of arsenic pollution treatment, as well as the existing relevant issues according to the change curves of the average concentration of arsenic in Yangzonghai Lake as published by Kunming Environmental Monitoring Center (KEMC) since 2008, were investigated.

2. Method

The polluted water area of Yangzonghai Lake is 31 km^2, and its water volume is 604 million m^3. Thus, the arsenic reduction project is extensive. However, no processes, such as pre-oxidation or stirring in engineering technologies, were conducted. The lake water can be directly used for preparing and diluting the $FeCl_3$ solution. Every day, the spraying ships are employed to carry out carpet spraying. Hence, the operation is very simple.

2.1. Spraying equipment

Ten tourist ships with a carrying capacity of approximately 5 t were remolded as spraying ships. On each ship, there were two polypropylene liquid-storage tanks with a volume of 1 m^3, a diesel engine, a suction sinking pump, and a spraying centrifugal pump so as to form a pipe network for adjusting the $FeCl_3$ solution concentration.

2.2. $FeCl_3$ solution preparation

Two polypropylene reactive tanks of 2 m^3 were used for dissolving $FeCl_3\cdot6H_2O$. Two polypropylene liquid-storage columns with a diameter of 2.5 m and a volume of 25 m^3 were used for storing concentrated $FeCl_3$ solution. In addition, several pumps and pipes, which can directly transport the stored solution to the liquid-storage tanks in the spraying ships, were installed.

2.3. $FeCl_3$ quality control

High concentrations of $FeCl_3$ liquid for the project were specially produced by Yunnan Salt & Salt Chemical Co., Ltd. In addition, two chemical factories were designated to supply $FeCl_3\cdot6H_2O$ products. The project also strictly stipulated the purity and content restrictions of harmful elements such as lead, cadmium, and mercury so as to ensure that there are no new harmful elements introduced to the water body during treatment.

2.4. Carpet spraying

Ten spraying ships were used for carpet spraying 6–8 t of $FeCl_3$ solution in the lake every day since mid-October 2009. At the initial stage, spraying was mainly concentrated in the southern water area. Later, this range was gradually expanded northwards. The spraying acreage covered four-fifth of the lake surface. After spraying the $FeCl_3$ solution into the lake, hydrolytic precipitation and arsenic adsorption occurred. Orange-red flocculent precipitates were observed by the naked eye to drift away from the sterns and gradually sink into the depths. Also, a large number of CO_2 bubbles were observed.

2.5. Water sampling system

Sampling was conducted each Wednesday and Saturday. A specific person was appointed in charge, who exactly was aware of the changes of the concentration of arsenic in the lake. According to the three sampling sites—southern site (N24°52′42.02″, E102°59′41.91″), central site (N24°54′17.66″, E102°59′58.07″), and northern site (N24°56′43.21″, E103°00′31.54″)—specified by KEMC,