



Eutrophication control using a novel bentonite humic-acid composite material BephosTM



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ABSTRACT

The main objective of the present study was to estimate the efficiency of BephosTM as an active capping agent preventing the phosphate release from eutrophic lake sediments. BephosTM was compared with unmodified bentonite (*N*-bentonite) and natural zeolite (*N*-zeolite), in a ratio 1/10/10 respectively. BephosTM as a P-inactivation agent resulted in about ~96.6% reduction of the phosphate flux and ~75.2% reduction of the ammonium flux from the sediments respectively. *N*-Zeolite and *N*-bentonite as capping materials resulted in about 64% and 91.8% reduction of the phosphorus flux and in about 70% and 35.6% reduction of the ammonium flux from the sediments respectively. Moreover, an additional purpose of this study was to investigate the different P-forms present in sediments, their contributions to the P-loadings of the ecosystem and also the effect of BephosTM amendment on P-binding properties of sediments. The results show that BephosTM restrained P release from the sediments, reducing the proportion of P-mobile in sediments, which was transformed into non-reactive species. The application of BephosTM caused an increase in the mass of P present in the more refractory 'apatite bound P' fraction compared to P-mobile, and also more P was stored in the residual P fraction of sediment post-application.

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Introduction

Humans' present existence and population growth are entirely based upon the development and efficiency of agriculture. Seafood delivers only a small percentage of human food world wide. A consequence of the increased population has been large-scale human-induced eutrophication. Cultural eutrophication has been considered as one of the major water pollution issues all over the world, resulting in harmful algal blooms and subsequent depletion of dissolved oxygen as well as alteration of ecosystem community structure [1–4].

Lake restoration efforts were traditionally focused on reducing nutrient inputs from the catchment, such as sewage discharges and diffuse runoff from agricultural land. Catchment remediation works including restricting stock access to the lake, enhancing riparian buffer zones and installing constructed wetlands on the main inflow have been previously used [5,6]. However, the in-lake

concentration was still high enough to maintain eutrophic conditions. Thus, even in cases where this action has been quite successful, the recovery of the waterbody may still be very slow due to internal loads. Phosphorus release from the sediment into the lake water (Fig. 1) may be so intense and persistent that it prevents any improvement of water quality for a considerable period after the loading reduction [7,8]. In order to reduce the release flux of nutrient from the contaminated sediments, various remediation strategies have been developed, such as dredging [9,10], precipitation of P by aluminum salts [11–13], in situ chemical injection [14], hypolimnetic oxygenation [15], and in situ capping [16]. Among these approaches, P-inactivation agents are used for reducing sediment nutrient release rate by permanently blocking the release of N and P from the sediments. In situ capping, which involves placement of a layer at the sediment–water interface to create a barrier between the sediment and the aquatic environment, is a potentially effective technology to stabilize sediments, minimize both re-suspension and transport, and reduce dissolved contaminant transport into overlying waters [1,17,18].

BephosTM (from the words “bentonite” and “phosphorus”) is a novel low-cost composite material embedding Fe, Cu ions and humic acid in the interlayer space of a natural bentonite. Its

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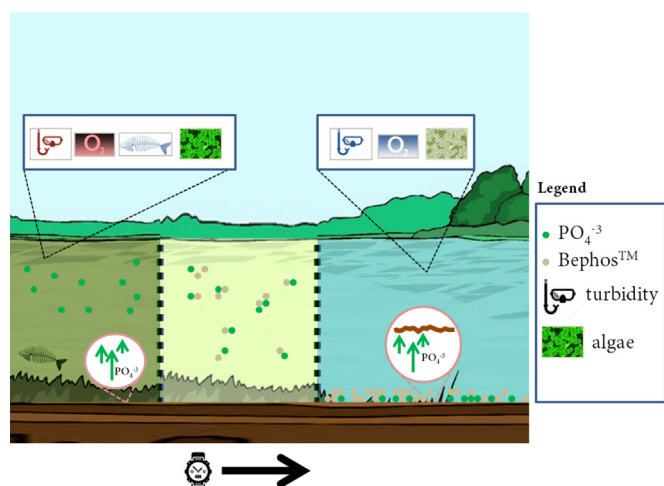


Fig. 1. Schematic approach of Bephos™ application in eutrophic water bodies.

properties have been studied in detail previously [19]. The modified clay was highly efficient for simultaneous phosphorus removal from water at a wide range of pH values (5–9). Its efficiency is maintained at alkaline pH (pH 8–9) in contrast with most materials, whose ability in adsorbing phosphorus drops dramatically under these conditions. This is of particular importance as in eutrophic waters the pH conditions that dominate are between 8 and 9. In addition, the adsorption capacity for phosphate and ammonium followed the order: fresh water > brackish > marine water.

The main objective of the present study was to investigate the efficiency of Bephos™ as an active capping agent in a pilot scale of real environmental conditions preventing the phosphate and ammonium release from anaerobic sediments. For this, Bephos™ was compared with unmodified bentonite (*N*-bentonite) and natural zeolite (*N*-zeolite). Moreover, different P-forms present in sediments, and their contributions to the P-loadings of the ecosystem, along with the effect of Bephos™ amendment on P-binding properties of sediments were investigated.

Materials and methods

Materials

The *N*-bentonite and *N*-zeolite samples used in this work were supplied by the company S&B from a mining site in Bulgaria. It is indicated that the analyzed natural zeolite contain 95% clinoptilolite ($\text{Ca Si}_7 \text{Al}_2 \text{O}_{18} \cdot 6\text{H}_2\text{O}$). Bephos™ was synthesized as it is reported in Zamparas et al. [19].

Sampling

During sampling, continuous profiles of physicochemical parameters such as temperature (*T*), conductivity (*C*), dissolved oxygen (DO), redox potential (*Eh*) and pH were measured in situ, using a Troll 9500 water quality multi-parameter instrument (Table 1). Environmental water and sediment samples were collected from the eutrophic Aitoliko Lagoon in July 2013 [20]. Sediment was collected at an approximate depth of 25 m using a bottom grab sampler. The samples were closed in air-sealed bags. Water sample for phosphate analysis was obtained from the water column 50 cm above the sediment sampling site. Water sample was collected, using a 2.5 L Hydro Bios free flow sampler. All the equipment used for sample collection, transportation, and preparation were free from phosphorus contamination. All the

Table 1

Physicochemical characteristics in the bottom layer of Aitoliko lagoon.

Parameters	
Temperature (°C)	13.61
Salinity (PSU)	25.48
pH	7.01
Dissolved oxygen (mg/L)	0
Redox potential (mV)	−335
Hydrogen sulfide (mg/L)	41.4
TP (mg/L)	0.507
PO_4^{3-} (mg/L)	0.424

samples were brought to the laboratory in a portable fridge at 4 °C within 2 h.

Phosphate sorption batch experiments

Adsorption kinetic data of phosphate on *N*-bentonite, *N*-zeolite and Bephos™ in contact times ranging between 15 and 480 min were studied using natural aqueous solution. 50 mg/L of natural water from Aitoliko lagoon, with initial phosphate concentration of 0.231 mg/L were added into conical flasks containing 0.1 g of each material. The suspension was separated by centrifugation and the concentration in supernatant (c_e) was measured by the molybdate blue spectrophotometric method. The amount adsorbed (q_e) was calculated from the difference in concentration between initial (c_o) and the equilibrium concentration. Blank samples without any adsorbent were perpetrated and monitored as a control. All experiments were carried out in duplicates.

P-partitioning in sediments

The determination of total phosphorus (TP) in sediments cannot predict the potential ecological danger related with eutrophication. Thus, the long term contribution of P bound in sediments can be more effectively evaluated on the basis of different P-fractions. P-fractions in sediments were extracted according to Hupfer et al. [24] (Fig. 2). The sum of labile P, reductant-soluble P and organic P fractions was defined as bio-available P (P mobile) and represent the potential release-sensitive P.

P in each fraction was calculated by:

$$P_{\text{sed}} = \frac{(P_f) \times V_{\text{extract}}}{\text{DW}} \quad (1)$$

In which P_{sed} is phosphorus in sediment ($\mu\text{g P g}^{-1} \text{ DW}$); P_f is phosphorus in fraction (mg P mL^{-1}); V_{extract} is the volume extractant (mL); DW is the dry weight sediment (g).

Column experiments

Sediment pH was found 7.2 and measured by a digital pH meter in 1:2.5 (W/V) sediment-to-water suspensions. Total phosphorus (TP) of the sediment was found 458 mg/kg P and measured by the molybdate blue spectrophotometric method. The sediment incubation experiments were carried out to estimate the efficiency of natural zeolite (*N*-zeolite), natural bentonite (*N*-bentonite), and modified bentonite Bephos™ as a sediment capping materials reducing the phosphate fluxes under anaerobic condition. Phosphate release measurements were conducted on well-mixed wet sediments collected from anoxic Aitoliko lagoon. The sediments (5 cm of thickness) were put in 4 glass cylinders (1 L) with an internal diameter of 4 cm; 700 mL of deionized water was added into each column. 10 g of *N*-bentonite and *N*-zeolite and also 1 g Bephos™ were carefully introduced into columns and one other acted as a control column simulating phosphate release from sediments under anoxic conditions. The columns were allowed to

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