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Management of arsenic-accumulated waste from constructed wetland treatment of mountain tap-water

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ABSTRACT

Arsenic-contaminated mountain tap water supply in Ron Phibun District, Nakorn Si Thammarat Province, Thailand poses a health hazard. Arsenic was removed using a constructed wetland (CW) system, in which the treated contaminated tap water was sedimented in 3 consecutive ponds before entering into the CW ponds, containing either *Cyperus* spp. or *Colocasia esculenta*. Following 1 year of operation both plants had similar ability to remove arsenic from mountain tap water. Arsenic was mostly concentrated at the roots of both plants. Arsenic in *C. esculenta* leaves was allowed to leach out in freshwater for 149 days, but the level (0.05 mg L^{-1}) was much lower than standard guidelines for industrial discharge. For *Cyperus* spp., young shoots were utilized as ornamental plants. As the sediments contained high arsenic levels, they were converted by a solidification/stabilization (S/S) system into cement-containing blocks, which after curing for 21 days produced arsenic leaching at levels that did not require a secure landfill for storage. The success of this study demonstrated that CW combined with appropriate S/S system is a suitable approach for Thailand in removing arsenic from contaminated water.

1. Introduction

Arsenic is a waste product from mining industry, in particular tin, and has resulted in contamination of the environment of many regions in Thailand. One such area is Ron Phibun District, Nakorn Si Thammarat Province in southern Thailand. The site is part of the Asian tin belt and has over a 100 year history of bedrock and alluvial mining. Consequently, soil and water in Ron Phibun District are grossly contaminated with arsenic. The water runoff from the mountain is directly distributed to the local population in Ron Phibun District via pipelines. In 1987, the Ministry of Public Health, Thailand, reported more than 1000 patients suffering from arsenicosis, known as "Blackfoot" disease in Thailand [1,2]. The arsenic contaminating surface water (0.50 mg L^{-1}) is much higher than World Health Organization (WHO) standard guidelines (0.01 mg L^{-1}) [3], and thus, Ron Phibun District was designated a high cancer risk area [2]. As arsenic in water cannot be detected by color, taste or smell, and cannot be biodegraded [4], it is imperative that the arsenic water level be reduced below accepted safety levels.

Designing an arsenic water treatment and management program is under a number of constraints, viz. financial, human resource and economic, so as to be able to achieve "more with less". So-called "appropriate" designs have been demonstrated throughout the world to be inoperable where there is an inability or unwillingness to impose limits [5,6]. The construction of artificial wetlands for treatment of metal contaminated water is now a widely accepted and increasingly common treatment alternative to high technology treatment [6]. Macrophytes are the main biological components of such constructed wetlands. They do not only assimilate pollutants directly into their tissues, but they also act as catalysts of detoxification reactions by increasing the environment diversity in root zones and by promoting a variety of chemical and biochemical reactions, which enhance the detoxification processes [7]. *Colocasia esculenta* and *Cyperus* spp. are the most widely used plants in constructed wetlands because of their fast growth rates and large uptake of nutrients and metal contaminants [8,9].

Therefore, we have compared the efficiency of *C. esculenta* and *Cyperus* spp. to remove arsenic from water in a constructed wetland system in Ron Phibun District. The trial was conducted for 1 year. The arsenic-accumulated products, viz. plants and sediments, were treated as hazardous wastes, using locally feasible management methods such as degrading in freshwater; marketing as ornamental plants or performing solidification/stabilization (S/S). Leaching test was conducted on S/S blocks in order to assess their suitability to be

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stored in secure landfills. Detoxification of arsenic accumulated in freshwater plant biomass was examined with *C. esculenta*, and the benefit of using arsenic-accumulated plants as ornamental plants was evaluated with *Cyperus* spp.

2. Materials and methods

2.1. Constructed wetland (CW)

2.1.1. Setting up CW

Free surface water CW was built as a series of ponds, into which the arsenic-contaminated water flowed before being discharged into either *Cyperus* or *C. esculenta* wetland pond. The test was performed at the arsenic polluted area in Ron Phibun District, Nakorn Si Thammarat Province, Thailand. The sedimentation pond, 2 m × 7 m × 2 m ($w \times l \times h$), was divided equally into 3 small ponds. The arsenic-contaminated tap water flowed into pond 1, and overflowed into pond 2 and then into pond 3. Suspended particles in the water were allowed to sediment to the bottom of these ponds. The wetland pond was a concrete pond, 2 m × 7 m × 1.8 m ($w \times l \times h$), filled with the following media layers: small gravel at the lowest level (10-cm thickness), sand in the next layer (10 cm) and 60-cm red soil mixed with ground coconut shell and commercial black soil (Sida soil) at the top. The efficiency of each wetland pond to remove arsenic from water was compared during 1 year of operation.

Young *Cyperus* spp. shoots were planted in 40 groups, 10–15 shoots per group, 30 cm apart. *C. esculenta* plants, 12 in. tall, were planted in 7 rows with 14 plants per row. After 6.5 months, as plants had overgrown, half of them were harvested by cutting off at the soil level. It took only 4 months for these plants to grow to the same height, 1.5 m, as the others. Phosphate fertilizer (N:P:K = 6:18:6) was added to the soil at planting and after the first harvest.

2.1.2. Operating CW

On each day arsenic-contaminated tap water from the sedimentation pond 3 was pumped into both of the CW ponds until the water level was 15 cm above the soil. Water was added to each CW pond in the morning (08.00 h) and drained out in the evening (17.00 h). The quantity of arsenic contaminated-water treated was 2100 L per pond per day. Water samples were collected from both the influent and effluent at weekly intervals.

2.2. Management of the arsenic-accumulated plants

2.2.1. Degradation in freshwater

C. esculenta leaves (50 g) were immersed for 149 days in 2 L of tap water kept outdoors. Water samples (30 mL) were collected 9 times from the start until the end of the experiment to determine the levels of arsenic released into the water. The non-As containing tap water was added to replace that removed. There were 3 replicates for each experiment.

2.2.2. Utilization as ornamental plant

As *Cyperus* plants in CW pond grew very fast and produced many young shoots, these were transferred to small pots as ornamental plants. Young shoots were harvested and utilized at the age of five months.

2.3. Management of arsenic accumulated soil and sediment

2.3.1. Determination of arsenic concentration in wetland soil and sediment

From each CW pond, 5 soil samples were collected at the level of 10–15 cm below the surface and were combined into 1 sample for arsenic determination. The collected soil samples were stored in sealed bags and kept at 25 °C until analysis.

Sediment from each of the 3 sedimentation ponds were dried in the sunlight for 7 days weighed and stored separately. A portion (125 g) of the samples from each site was heated in an oven at 60 °C for 24 h, homogenized by hand, passed through a 2 mm mesh size sieve and then ground (mortar/pestle) to a powder of 280 μm in size.

Determination of arsenic was performed in a hydride generation atomic absorption spectrometry (HG-AAS).

2.3.2. S/S procedure

The ratio of solid material to water used in S/S procedure was 1:0.42. Solid material used as control in the study was Portland cement mixed with sand at the ratio 1:3. The solid material was modified (as treatment) by combining with 25% sediment. After the solid material was mixed with water, it was allowed to harden as blocks, 3.5 cm × 7.0 cm, and cured at room temperature (25 °C) in plastic bags. To determine whether the curing time had influenced the amount of arsenic leaching out from the S/S blocks, samples were removed at 14 and 21 days of curing and subjected for a leaching test using the Toxicity Characteristic Leaching Procedure (TCLP) Method 1311 [10]. The leaching experiment was also performed using deionized (DI) water as leachant in order to simulate a scenario in which S/S block is in contact with rainwater.

2.3.3. Leaching test

TCLP method was performed according to US.EPA Method 1311 [10]. Five grams of treated and untreated samples, particle size of less than 5 mm, were extracted with acetic acid solution (5.70 mL of glacial acetic acid diluted with reagent water to a volume of 1 L, pH 2.88 for treated samples; and 5.70 mL of glacial acetic acid diluted to 500 mL of reagent water mixed with 64.3 mL of 1 N NaOH, and diluted to a volume of 1 L with reagent water, pH 4.93 for untreated samples) at a liquid:solid ratio of 20:1. Acetic acid solution (0.10 M, pH 2.88) was used to extract cement-treated samples due to the high alkalinity of the wastes. The solid and leachate suspension were placed in a capped polypropylene bottle and shaken at 30 rpm for 18 h. After the extraction, the pH of the leachate was measured, and the liquid was separated by filtration through a 0.45 μm membrane fiber filter. The soluble arsenic concentration in the filtrate was analyzed with HG-AAS.

For DI water extraction, the procedure was the same as with TCLP except that DI water was used as leachant. Leachates were collected at curing times of 14 and 21 days.

2.4. Total arsenic analysis

2.4.1. Plant samples

Plants were harvested, washed with tap water and rinsed with DI water. They were then separated into various parts and oven-dried at 50–55 °C for 3 days. The weights of dry biomass of each plant part were determined before being ground to powder with mortar and pestle. Samples were prepared for arsenic concentration analysis using the dry ashing method [8]. A portion of the dry powder (~20–50 mg) was accurately weighed (±0.01 mg) directly in a crucible and mixed with 1.5 mL of freshly prepared slurry (30 g of Mg $(NO_3)_2$, 50 g of MgO and 500 mL of DI water). Mixture was dried overnight at 80 °C, and heated in a muffle furnace (200 °C for 1 h, 300 °C for 1 h and 500 °C for 8 h). Residue was dissolved in 2.5 mL of 6 M HCl and then 2.5 mL of distilled water were added. Total arsenic concentration was determined by HGAAS using a Perkin-Elmer MHS-20 mercury/hydride system coupled to a Perkin-Elmer 2380 AAS. Arsenic concentration is reported on a dry mass basis. A certified reference material (MESS-3 marine sediment; National Research Council Canada, Ottawa, Canada) was used for quality control at 85 ± 7% recovery.

Table 1
Arsenic concentration in plant parts (mg kg^{-1}).

Plant part	Arsenic concentration (mg kg^{-1})	
	Cyperus spp.	*C. esculenta*
Root	190 ± 55.0	602 ± 32.1
Bulb	–	7.40 ± 3.35
Stem	2.98 ± 1.46	5.34 ± 1.22
Leave	5.16 ± 1.74	4.63 ± 1.74

Table 2
Mass balance of constructed wetland plants.

	Cyperus spp.	*C. esculenta*
Plant growth (g m^{-2} d^{-1})	1597	20.3
Plant uptake (g d^{-1})	4.41	0.17
Accumulated soil (g d^{-1})	15.2	18.4
Influent water (g d^{-1})	0.92	0.92
Effluent water (g d^{-1})	0.21	0.20
Percent arsenic removal	77.0	78.7

2.4.2. *Soil and sediment samples*

Five hundred g of sample were dried at 60 °C for 12 h, and then 125 g was sieved through a 2 mm mesh in order to remove stones and plant material. Each sample was then ground using a mortar and pestle and passed through a 280 μm sieve before being analyzed for arsenic content by HGAAS.

2.4.3. *Water samples*

Arsenic concentrations in water samples were determined directly without digestion of the acidified samples after filtering through a 45 μm membrane filter.

3. Results

3.1. *Efficiency of wetland plants in arsenic removal*

Arsenic-contaminated tap water has been provided for people in Ron Phibun District for more than 10 years [11]. In order to reduce the arsenic content in tap water, a remediation procedure was established in which tap water was passed through a series of three sedimentation ponds and two CW ponds. After 1 year of operating the CW ponds, both *Cyperus* spp. and *C. esculenta* showed similar efficiency in removing arsenic from an initial level of 0.36 mg L^{-1} in the influent down to 0.08 mg L^{-1} (22%) in the effluent water. The quantity of tap water processed by each CW system was 885 L per day. After 1 year, the average number of *Cyperus* spp. had increased from 12 to 114 plants per group of the 40 groups. Thus, 0.19 L of arsenic-contaminated water was treated by each *Cyperus* plant per day, while for *C. esculenta* it was 9 L per plant per day.

Arsenic was mostly accumulated at the roots of wetland plants, 602 mg kg^{-1} and 190 mg kg^{-1} for *C. esculenta* and *Cyperus* spp., respectively (Table 1). The average growth of *Cyperus* spp. was 1597 g m^{-2} d^{-1} compared with 20 g m^{-2} d^{-1} of *C. esculenta* (Table 2). Similarly, arsenic uptake by *Cyperus* spp. (4.41 g d^{-1}) was 26 times higher than that of the *C. esculenta* (0.17 g d^{-1}). Stem height and root length of *Cyperus* spp. was in the range of 190–200 cm and 20–28 cm, respectively; dry weight biomass was 1.69 kg per plant and the increase in plant number per group was 105–125 plants as counted at harvest on 345 day. Stem height and root length of *C. esculenta* was 135–150 cm and 10–20 cm, respectively, and dry weight biomass was 1 kg per plant.

3.2. *Management of arsenic-accumulated plants*

3.2.1. *Leaching by freshwater*

Arsenic leached by tap water (2 L) from leaves and petioles of *C. esculenta* (50 g) were monitored over a period of 149 days. At the start of the experiment (t_0), the average arsenic concentration in the leaves of *C. esculenta* was 1.80 mg kg^{-1}, and the amount of arsenic leached out from the plant biomass into water was in the range of 0.01–0.09 mg L^{-1} (Fig. 1). The highest arsenic concentration leached out was at day 9, and then gradually declined until the end of the test period. Plant biomass was not completely leached out.

3.2.2. *Utilizing Cyperus spp. as ornamental plants*

Cyperus spp. plants grew very fast in the CW pond and produced many young shoots, which were transferred into small pots for further growth as ornamental plants. A pot of 10–15 plants, 60–90 cm in height, was worth 80–120 baht (∼3–4 US$) at the local market.

3.3. *Management of arsenic-accumulated soil and sediment*

3.3.1. *Arsenic concentration in soil and sediment*

Arsenic concentration in soil samples from CW ponds was 35.80 ± 6 and 41.17 ± 15 mg kg^{-1} for *C. esculenta* and *Cyperus* spp., respectively, at the start of the experiment. After 1 year, arsenic level in soil increased to 90 mg kg^{-1} for *C. esculenta* and 75 mg kg^{-1} for *Cyperus* spp. (Fig. 2). This level of arsenic in both soils was higher than Thailand soil standard of 3.9 mg kg^{-1} for agricultural land and 27 mg kg^{-1} for non-agricultural land [12].

After 3 months operating the CW, sediment (S) was collected from the 3 sedimentation ponds and arsenic contents determined. The highest arsenic level was found in the sediment of pond 1 (23,438 mg kg^{-1}), with decreased levels in ponds 2 (37%) and 3 (61%) (Fig. 3).

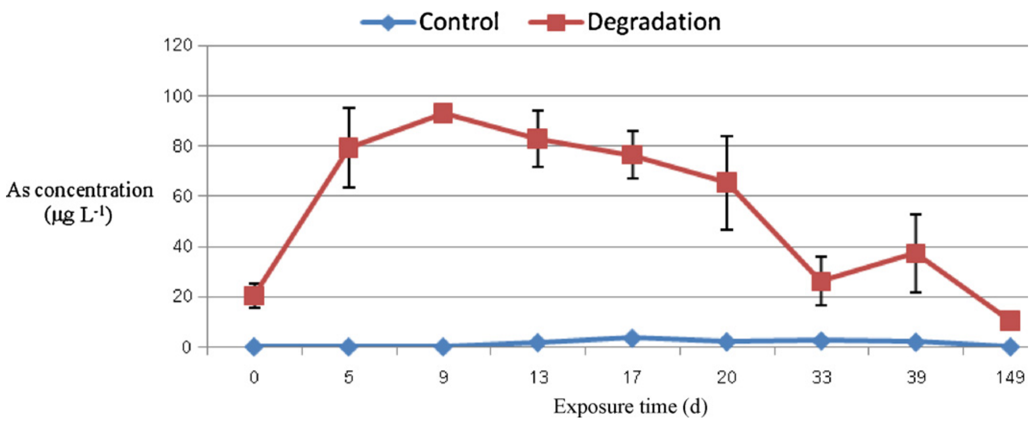

Fig. 1. Arsenic concentration (μg L^{-1}) in the water during degradation in freshwater.

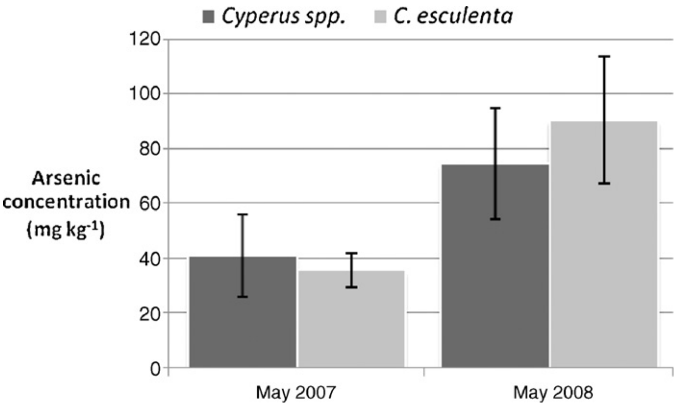

Fig. 2. Arsenic concentration (mg kg^{-1}) increased in the wetland soils during 1 year experiment.

3.3.2. *Arsenic leaching of untreated waste*

Untreated wastes were determined for the amount of arsenic leached out by TCLP and DI procedures. TCLP was more effective than DI in leaching arsenic (Table 3). Arsenic leached out from untreated CW soil and sediment 1 was 0.02 and 0.09 mg L^{-1}, respectively. Thus, only sediment 1 was considered as a hazardous waste and was further managed by S/S treatment.

3.3.3. *S/S treatment*

There were 2 samples subjected to S/S treatment, cement-sand mixture (CSM) as control and CSM (75%) + sediment (S) (25%). At day 21, total arsenic concentration was 6.90 and 579 mg kg $^{-1}$ in control and treated sample, respectively. Arsenic leaching out from S/S blocks after 14 days of curing by TCLP was 0.01 and 0.73 mg L^{-1} in control and treated S/S sample, respectively (Table 4). After 21 days of curing, a lower total arsenic concentration was leached out by TCLP. However, the concentrations of arsenic leached out at both 14 and 21 days are significantly higher using TCLP than DI.

4. Discussion

In any arsenic phyto-remediation program, the issue of disposal of the contaminated biomass must be addressed because it is the most important hurdle in the implementation of the phytoremediation technology. A waste management hierarchy based on the most environmentally sound criteria favors waste prevention/minimization, waste re-use, recycling, and composting [5,13]. In developed countries, the main disposal methods are land filling and incineration [13]. However, in many developing countries, due to lack of resources, the construction of artificial wetlands for treatment of metal contaminated water is widely accepted as an alternative procedure [6].

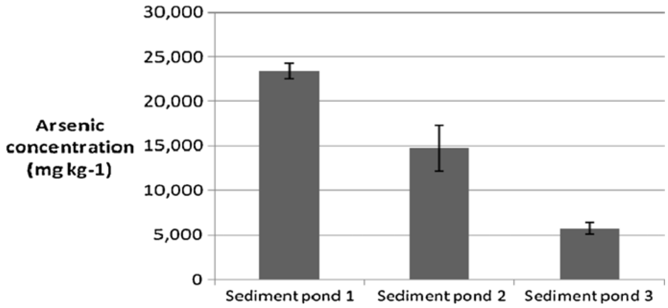

Fig. 3. Arsenic concentrations (mg kg^{-1}) of sediments in the sedimentation ponds.

Table 3

Leaching test of untreated waste from soil and sediment.

Sample	Arsenic (mg kg^{-1})	pH	Arsenic (mg L^{-1})
TCLP			
Cyperus soil	74.7 ± 20.2	4.90 ± 0.01	0.02 ± 0.004
C. esculenta soil	90.4 ± 23.3	4.90 ± 0.04	0.02 ± 0.003
Sediment no. 1	5069 ± 282	6.10 ± 0.19	0.09 ± 0.009
DI			
Cyperus soil		9.80 ± 0.93	0.003 ± 0.0003
C. esculenta soil		8.60 ± 0.10	0.003 ± 0.0002
Sediment no. 1		9.10 ± 0.32	0.050 ± 0.004

TCLP: toxicity characteristic leaching procedure; DI: deionized water procedure.

However, wetland systems produce considerable amounts of sludge and sediment and convert a water-based pollution problem to that of solid waste management. Treatment and disposal of sewage solids (sludge) are major components in the overall treatment costs, and should therefore be taken into consideration. A particular problem arising from the disposal of treatment residues generated by removal of arsenic from drinking water is that arsenic can be highly mobile and has the potential to leach back to ground and surface waters [14]. In predicting the ecological impact of trace metals, estimation of their potential leachability, i.e., the fraction of total metal content that may become leachable is more relevant than the total metal analysis.

S/S includes a range of processes, which are normally used as a pre-landfill waste treatment that aims to make hazardous wastes safe for disposal [15]. S/S process involves mixing the waste, either in the form of sludge, liquid or solid, into a cement-binder system. S/S is most suitable for treating wastes that are predominantly inorganic, as these are considered more compatible with the cement binders used. S/S inhibits leaching of hazardous components by reducing waste/leachant contact and by forming a stable pH environment in which many heavy metals of environmental concern remain insoluble [16]. Many studies have shown that industrial waste containing arsenic (III) could be successfully treated using S/S technology employing lime and cement [17–20]. Akhter et al. [21] found that Portland cement is the best binder in the stabilization of soils contaminated with heavy metals and 10,000–12,200 mg L^{-1} sodium arsenite. Portland cement has been successfully used to stabilize arsenic-rich sludge [14]. The results of using S/S in preventing arsenic leaching showed that arsenic level of the treatment was lower than the Maximum Contaminant Level. Choi et al. [16]

Table 4

Leaching test for S/S treated waste.

Sample	pH	Arsenic (mg L^{-1})
d 14		
TCLP		
Formula 1[a]	5.60 ± 0.06	0.01 ± 0.001
Formula 2[b]	5.50 ± 0.12	0.73 ± 0.002
DI		
Formula 1[a]	12.4 ± 0.06	ND
Formula 2[b]	12.6 ± 0.10	0.004 ± 0.001
d 21		
TCLP		
Formula 1[a]	5.50 ± 0.00	0.007 ± 0.001
Formula 2[b]	5.40 ± 0.06	0.61 ± 0.02
DI		
Formula 1[a]	12.4 ± 0.15	ND
Formula 2[b]	12.5 ± 0.12	0.002 ± 0.001

d: day; TCLP: toxicity characteristic leaching procedure; DI: deionized water procedure; ND: not detected.

[a] Formula 1: CSM 100% = 6.90 ± 1.86 mg kg^{-1}.

[b] Formula 2: CSM 75% + S 25% = 579 ± 206 mg kg^{-1}.

found that 7.5% addition of cement is optimum in reducing arsenic leachability. The important mechanism to immobilize arsenic in the cement-based S/S is the precipitation of $Ca_3(AsO_4)_2$ and the adsorption to cement hydrates [16].

In this study, the surface flow CW using either *C. esculenta* or *Cyperus* spp., locally grown wetland plants, was selected to assess their appropriateness in removing of arsenic from tap water in Ron Phibun District, Nakorn Si Thammarat Province, Thailand [9]. The retention time used in the study was 9 h, which was shown to be sufficient to remove arsenic from the influent water and the effluent could be safely discharged into a natural reservoir. Jenssen et al. [7] have used CW to remove divalent metals, such as copper, lead, and zinc, and more recently Nemade et al. [22] employed constructed soil filter that results in oxidation of As (III) to As (V) and co-precipitation by iron salt and removal of arsenic below WHO standard. Despite the high efficiency of the wetland system to remove arsenic, the wastes must be treated as hazardous as they accumulated the toxic metal. Young shoots of *Cyperus* spp. could be harvested and further grown as ornamental plants.

5. Conclusion

Removing arsenic from the contaminated water by the CW system was shown to be very successful. The arsenic levels in the effluents were below safety standards and could be able to be discharged into the natural environment. Economic gain and public acceptance are important criteria for the success of any technology to be applied for remediation. This study showed that CW using *Cyperus* spp. could not only remove arsenic from contaminated water but could produce income as ornamental plants. Management of arsenic-accumulated plants, which has been a hurdle of the implementation of phyto-remediation, was successfully solved using S/S technology employing a cement binder, which produced leached arsenic levels lower than Thailand's maximum contaminant level of $5\,mg\,L^{-1}$, and thus the S/S blocks did not require a secure landfill for storage. This is critical to the success of a solid waste management system particularly in a low-technology economy country.

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