J. Eng. Technol. Sci., Vol. 52, No. 1, 2020, 66-80

Phytoremediation of Mercury Contaminated Soil with the Addition of Compost

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Highlights:

- The reactor with growth media consisting of 80% contaminated soil and 20% compost with *Sansevieria trifasciata* had 75.63% Hg removal efficiency with the final Hg concentration at 58 mg/kg.
- The reactor with growth media consisting of 80% contaminated soil and 20% compost with *Celosia plumosa* had 66.81% Hg removal efficiency with the final Hg concentration at 79 mg/kg.
- The reactor with growth media consisting of 100% contaminated soil with *Sansevieria trifasciata* had 74.79% Hg removal efficiency with the final Hg concentration at 60 mg/kg.
- The reactor with growth media consisting of 100% contaminated soil with *Celosia plumosa* had 65.55% Hg removal efficiency with the final Hg concentration at 82 mg/kg.

Abstract. Phytoremediation is an alternative technology for processing mercury (Hg) contaminated soil. The objective of this study was to treat Hg contaminated soil by adding compost using *Sansevieria trifasciata* and *Celosia plumosa*. The variations of the composition of the growth media were 100% contaminated soil and 80% contaminated soil with 20% compost. The plants used were aged 1 month and 30 cm high. The reactor was a polybag with a diameter and height of 25 cm. Sampling was conducted once every 7 days for 28 days. This research showed that the reactor with growth media consisting of 80% contaminated soil and 20% compost with *Sansevieria trifasciata* and *Celosia plumosa* had an Hg removal efficiency of 75.63% (58 mg/kg) and 66.81% (79 mg/kg), respectively. The Hg removal efficiency with growth media consisting of 100% contaminated soil was 74.79% (60 mg/kg) and 65.55% (82 mg/kg) in the reactor with *Sansevieria trifasciata* and *Celosia plumosa* respectively.

Keywords: *Celosia plumosa; compost; mercury; phytoremediation; Sansevieria trifasciata*

1 Introduction

Small-scale gold mining (ASGM) is one of the mining practices in Indonesia. Mercury-dependent artisanal ASGM is the largest source of mercury pollution on

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Received July 31st, 2019, Revised November 6th, 2019, Accepted for publication January 27th, 2020.
ISSN: 2337-5779,
DOI: 10.5614/j.eng.technol.sci.2020.52.1.5

earth [1]. Mercury amalgamation is a traditional method used in small-scale gold mining to recover gold [1-2]. The processing of gold ore by amalgamation uses mercury as a gold binder [1]. The gold is separated via a cyanide process until gold and silver alloys are acquired, while the mercury evaporates [3]. In the amalgamation process, an estimated 25-30% of the mercury used is released into the environment [4]. Oh, *et al.* [3] state that for each gram of gold produced, about 1 to 3 grams of mercury is released into the environment resulting from the amalgamation process. Mercury pollution occurs because of residual tailings that are scattered during removal and when the tailing storage pond is full [4].

The remaining traditional processes of mercury-contaminated waste and various other heavy metals in the form of sludge are disposed in agricultural land, which can have a negative impact on crop production [2]. Mercury can harm the environment because it is absorbed by plants quickly and endangers human life through the food chain [5]. The Minamata disease that has plagued Minamata city, Japan since 1985 originates from industrial waste containing mercury, has caused poisoning, nerve paralysis and hundreds of deaths [6]. Mercury is the only metal that has biomagnification through the food chain and can very easily transform into a more toxic organic form [7].

The control of mercury pollution is a worldwide commitment. The United Nations Environmental Program (UNEP) initiated a conference attended by the representatives of the Intergovernmental Negotiating Committees from each country to discuss and negotiate a global agreement on mercury. The conference produced the Minamata Convention on Mercury signed by 128 countries and the European Union [8]. This convention is a legal international instrument concerning global mercury management.

The goverment of the Republic of Indonesia committed to reducing and eliminating mercury pollution with the issuance of Law No. 11/2017 on the ratification of the Minamata Convention on Mercury [9]. Its aim is to stop the use of mercury in small-scale gold mining and strengthen the regional capability to solve the problem of mercury pollution from small-scale gold mining. The approach taken includes searching for alternative mercury processing techniques.

Phytoremediation is an alternative technology to cultivate land contaminated with heavy metals [10]. It has several advantages, such as low operational costs, simple technology, and high efficiency in removing heavy metal from soil [2]. Phytoremediation is a process that uses various types of plants to remediate polluted land, removing, stabilizing and absorbing contaminants in the form of organic and inorganic compounds in the soil [10-11]. Phytoremediation uses the ability of various types of plants to accumulate or absorb heavy metals or organic compounds (phytoaccumulation) based on the characteristics of the plants to remediate polluted soils [3,11].

In a previous study, phytoremediation of mercury-contaminated soil using *Vetiveria zizanioides* could reduce the largest amount of mercury (8 mg Hg^{2+}/kg) in media consisting of 100% contaminated soil and 90% of contaminated soil : 10% compost [12]. The mercury reduction in the soil was in the range of 30 to 65% from an initial mercury concentration of 0.87 mg/kg after 28 days of exposure. Juhriah and Alam [13] concluded that *Celosia plumosa* could reduce the mercury concentration with a removal efficiency of 81.25 to 98.68%.

The study by Ulimma, *et al.* [14] using *Sansevieria trifasciata* could reduce the concentration of mercury in tailings media with a removal efficiency of 46.72% with a concentration of 0.55 mg/kg and in mixed media with a removal efficiency of 57.36% with a concentration of 0.38 mg/kg.

Few studies have investigated the use of phytoremediation with composting addition for the treatment of metal-contaminated soils. Related studies considered the remediation of oil-contaminated soil and oily sludge using phytoremediation and composting [15-16]. Another study considered using combinations of compost and plants from a different perspective [17]. The objective of this study was to treat Hg contaminated soil with compost addition using *Sansevieria trifasciata* and *Celosia plumosa*.

2 Materials and Methods

2.1 Characteristics of the Growth Media

The initial characteristics of the growth media were investigated to determine the concentration of mercury in the growth media. The experiment was done by using soil mixed with artificial waste in the form of Hg^{2+} standard solution that was deposited for 24 hours. The soil samples were dried, crashed, and sieved using a 0.5-mm sieve. After weighing 1 gram and putting it in an Erlenmeyer flask, 5 mL HNO_3 and 0.5 mL $HClO_4$ were added. The soil sample was then heated, left overnight and filtered. The filter results were analyzed using the atomic absorption spectrophotometer method [18].

In this study, the initial mercury concentrations in the artificial waste before being mixed into the soil were 150 mg/kg, 250 mg/kg, and 350 mg/kg. After the artificial waste was mixed into the soil, the mercury concentration was 143 mg/kg, 238 mg/kg, and 331 mg/kg respectively (Table 1). The mercury concentration in this soil exceeded the required quality standard of 0.05 to 2 mg/kg [19-21].

Table 1 Initial soil characteristics.

Initial concentration (mg/kg)	Soil concentration (mg/kg)	Quality standard for heavy metals Hg on the ground (mg/kg)* [20-22]
150	143	0.05-2
250	238	
350	331	

2.2 Acclimatization of Plant and Range Finding Test

This study used 1-month old *Sansevieria trifasciata* and *Celosia plumosa* plants with a height of 30 cm. Acclimatization of the plants was done for 1 week to make the plants adapt to the new environment [22]. The plants that were used in the next stage were fertile plants and did not experience death.

A range finding test (RFT) was conducted to determine the maximum concentration of mercury that could be tolerated by the plants [22]. The concentrations used were 150 mg/kg, 250 mg/kg, and 350 mg/kg. The RFT was conducted for 7 days. The concentrations that were still tolerable by the plants were used for phytoremediation of mercury-contaminated soil.

2.3 Phytoremediation of Mercury Contaminated Soil

This study was conducted in a laboratory by testing the ability of *Sansevieria trifasciata* and *Celosia plumosa* to remediate mercury in soil, and to see by how much they were able to decrease of the mercury concentration in the soil. The following variations of the composition of the growth media were used: (1) 100% contaminated soil, (2) 80% contaminated soil and 20% compost. Compost derived from an unstable compost of high organic material content was used as a stimulant in the process of phytoremediation [23]. The mercury concentration used refers to the results of the RFT, i.e. 150 mg/kg and 250 mg/kg. The study was done in duplicate, so 16 reactors were required (Table 2).

Table 2 Experimental conditions.

Reactor	Composition of planting media (contaminated soil:compost)	Hg concentrations (mg/kg)	Plants used
Reactor 1	100:0	150	*Sansevieria trifasciata*
Reactor 2	80:20		
Reactor 3	100:0	250	
Reactor 4	80:20		
Reactor 5	100:0	150	*Celosia plumosa*
Reactor 6	80:20		
Reactor 7	100:0	250	
Reactor 8	80:20		

The reactors in the form of a polybag containing soil were given a pollutant load with a predetermined mercury concentration. The polybag used had a diameter of 25 cm and a height of 25 cm. The growth medium used was 2 kg for each reactor. The parameters tested in this study were the mercury concentration in the soil, the pH value, and the soil temperature. Observation and soil sampling of the absorption of mercury were conducted every 7 days for 28 days and analyzed using the AAS method. Sampling was done using a small iron pipe with a hole and a spoon with a hole. The pipe was stuck into the soil until it reached the bottom of the polybag, after which the pipe was pulled out. The soil that was captured by the pipe was then put into a plastic cup. The soil sample collected in a plastic cup was put into a plastic bag to analyze the mercury level [24].

3 Results and Discussion

3.1 Acclimatization of Plants and RFT Result

Acclimatization of the plants was done to let the plants adapt to the new environment. Physical observations were carried out on both types of plants for a week. From the observations that were done, it could be seen that both types of plants experienced physical changes. From day 1 to day 2 the plants continued to flourish and were fresh and when entering the 5th day both types of plants experienced growth of new shoots until the 7th day. During the RFT phase, physical observations were done on both types of plants every day for a week. At the RFT stage, it was found that both types of plants could survive and accept mercury pollutant loads at concentrations of 0.143 mg/kg and 0.238 mg/kg. Although *Celosia plumosa* experienced a slight wilt in its leaves, it could still survive until the 7th day. Meanwhile, at an Hg concentration of 0.331 mg/kg both types of plants underwent physical changes, i.e. the leaves withered and dried. Hence, the concentration of 0.331 mg/kg was not used in the stage of the main research because this high pollutant load caused both plants to die.

According to Siahaan, *et al*. [25], chlorosis (yellowing) is the main symptom of plants with mercury poisoning. Mercury poisoning also causes the plant roots to turn brown, the number of leaves and root size to decrease, and the root caps to sustain damage. Mercury poisoning in plants can be caused by changes in cell membrane permeability, sulphydryl (-SH) group reactions with cations, affinity for reacting with active phosphate and ADP or ATP groups, and replacement of essential elements, especially macro elements.

3.2 PH Value and Temperature in Growth Media

The pH value in the growth media planted with *Sansevieria trifasciata* and *Celosia plumosa* during the study ranged between 6.5 and 7.0. The increase in pH in the growth media allegedly occurred because of an increase in the use of CO_2 in the process of photosynthesis [26]. Neutral soil (pH 6.0 to 7.0) is becomes to reach a nutrient equilibrium. Generally, the pH in the soil in the experiments ranged between 3.0 and 9.0. This shows that the pH of the growth media with *Sansevieria trifasciata* and *Celosia plumosa* was optimal for growth and productivity of plants and microorganisms in the soil [13].

The temperature values contained in the growth media planted with *Sansevieria trifasciata* and *Celosia plumosa* experienced changes from day 0 to day 28. The average temperature in the growth media in all reactors was within the range of the mesophilic temperature, i.e. around 25 °C to 30 °C. Mangkoedihardjo, *et al.* [26] state that land recovery can be done biologically in the mesophilic temperature range from around 25 °C to 40 °C. This shows that the temperature in the growth media was the maximum temperature at which plants can still grow and microorganisms in the soil are still able to develop to help the process of soil fertilization for plant growth.

3.3 The Decrease of Mercury Concentration in Growth Medium using *Sansevieria trifasciata*

The concentration of mercury in the growth medium was observed, as shown in Figure 1. The mercury concentration was adsorbed in the growth medium starting from the first day after exposure and fluctuated thereafter. The highest adsoprtion occurred on day 28 for almost all treatments. The mercury concentration in the growth medium showed a decreasing trend from day 7 to day 14 and then fluctuated until the end of observation. The results show that the mercury adsorption by the growth medium started to increase on day 21 and started to decrease on day 28. It can be seen that at the highest mercury concentration (238 mg/kg), the mercury removal from the growth medium was faster on the same day of observation. The highest mercury adsorption reached by the 100% contaminated soil was 71 mg/kg in the 143 mg/kg treatment. In the 238 mg/kg treatment, the mercury adsorption by the growth medium reached 60 mg/kg.

Decreasing the mercury concentration can be done by the processes of phytoremediation, namely phytoextraction, phytostabilization, rhizofiltration, phytodegradation, and phytovolatilization [10]. Phytoextraction is the absorption and translocation of contaminants by plant roots into various plant organs [12]. Phytostabilization means that the roots of a plant immobilize pollutants by accumulating and adsorbing them in the surface of the soil and precipitating pollutants in the root zone. Rhizofiltration means that the root of a plant adsorbs pollutants in the root zone using symbiosis between plants and microorganisms in the media around the roots [5]. Phytodegradation is the breakdown of contaminants that are absorbed through metabolic processes in plants. The mercury concentration in the soil also decreases because mercury is a type of heavy metal that can evaporate into the atmosphere. Mercury pollutants from the soil absorbed by *Sansevieria trifasciata* and *Celosia plumosa* are transformed and released in the form of liquid vapor into the atmosphere and absorbed by plant leaves. This process is referred to as phytovolatilization.

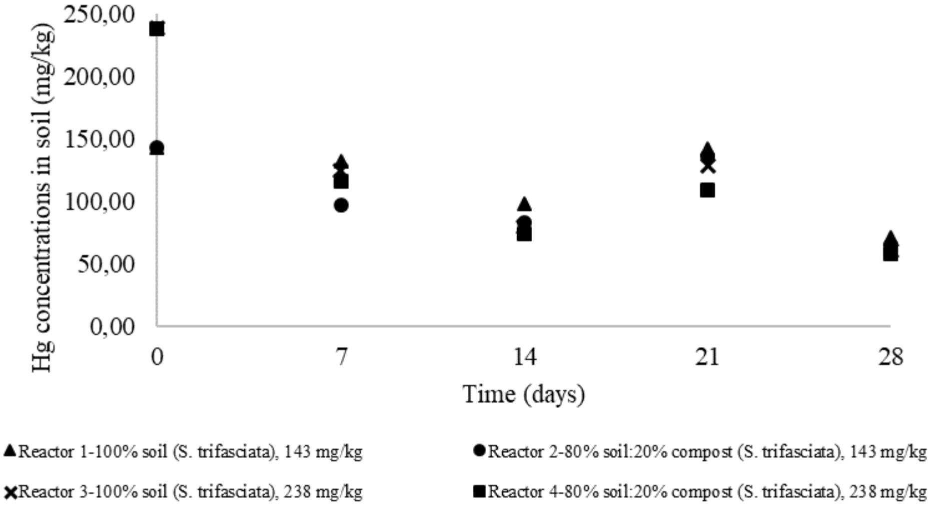

Figure 1 The decrease of the mercury concentration in growth medium using *Sansevieria trifasciata*.

The decrease of the mercury concentration in the growth media by the addition of compost (80% contaminated soil and 20% compost) was faster on the same day of observation compared to without addition of compost. The highest mercury concentration decrease happened on the 28th day. For the 143 m/L and 238 mg/L treatments, the mercury adsorption by the growth media reached 65 mg/L and 58 mg/L, respectively. According to Mangkoedihardjo, *et al.* [26] and Mangkoedihardjo and Triastuti [12], compost as a stimulant can help and improve the phytoremediation process. Organic material can be a source of carbon for microorganisms to stimulate the plant roots. The right combination of soil and compost increases the biodegradability of contaminated soils, and also the anion content of the compost has good absorption capacity so it can bind metal cations in the soil [23].

In all treatments, the mercury concentration decreased from the first observation on day 7 until the end of observation (Figure 2). For the 143 mg/kg treatment of 100% contaminated soil, the mercury removal efficiency was 50.35% on day 28. A similar trend occured in the treatment with 238 mg/L, where the mercury removal efficiency was 74.79% on day 28. For the growth medium using compost (80% contaminated soil and 20% compost) in the 143 mg/kg and 238 mg/kg treatments, the mercury removal efficiency was 54.55% and 75.63% on day 28, respectively. In the 238 mg/kg treatment, the growth medium could absorb more mercury than in the 143 mg/kg treatment. This is because the higher the amount of mercury concentration in the soil, the greater the amount of absorbed mercury. Compost addition has the impact of the plants improving the phytoremediation process, reducing the mercury concentration with a fairly high percentage [22]. Microorganisms can survive well and help the process of plant growth and enhance the phytoremediation process in the absorption of mercury in soil [12].

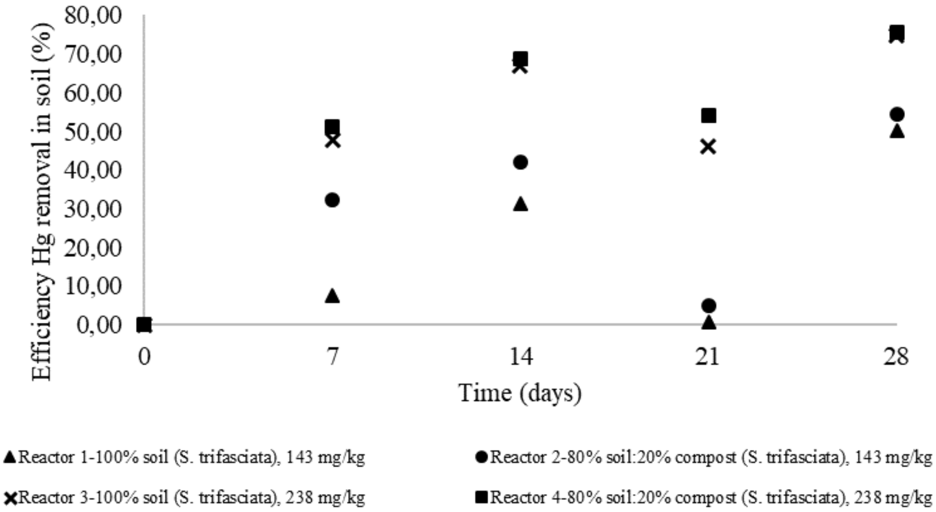

Figure 2 The mercury removal efficiency in growth medium using *Sansevieria trifasciata.*

3.4 The Decrease of Mercury Concentration in Growth Medium *Celosia plumosa*

The mercury was adsorbed by the growth medium starting on the first day after exposure and then decreased (Figure 3). The highest adsorption occurred on day 28 in almost all treatments. The mercury concentration in growth medium for 100% contaminated soil decreased from day 7 to the end of observation. For the 143 mg/kg treatment, the mercury concentration decreased by 68 mg/L on day 14 and 87 mg/L on day 28. In the 238 mg/kg treatment, the mercury concentration decreased by 102 mg/kg on day 14 and 82 mg/kg on day 28. A similar trend occurred in the treatment with 80% contaminated soil and 20% compost, with the mercury concentration decreasing by 73 mg/kg on day 14 and 67 mg/kg on day

28. In the 238 mg/kg treatment, the mercury concentration decreased by 91 mg/kg on day 14 and 79 mg/kg on day 28.

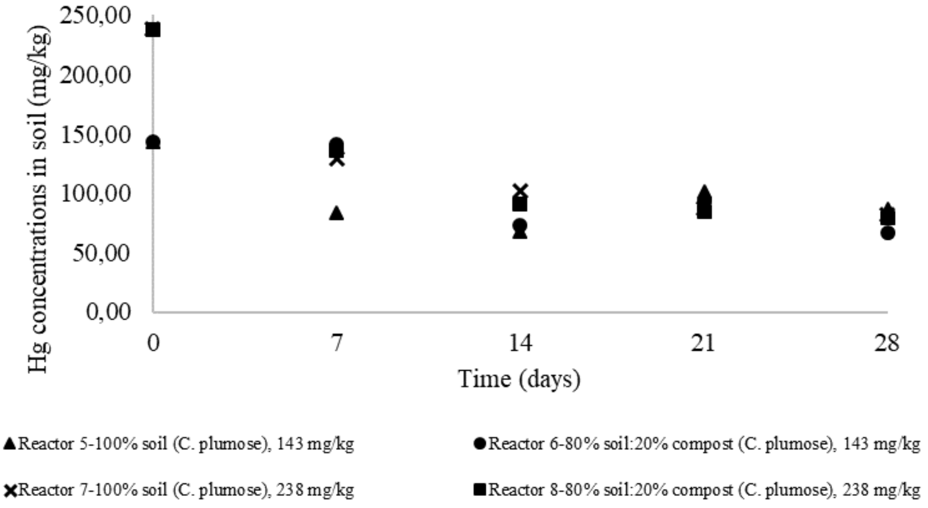

Figure 3 The mercury concentration decrease in growth media using Celosia plumose.

Heavy metal contaminants can be removed through several wetland system mechanisms [11,27]. These processes include sedimentation, microbial degradation, precipitaton, and plant uptake, removing most contaminants. Metals may be adsorbed to the soil or sediment, or may be chemated or complexed with organic matter. They can also precipitate out as sulfides and carbonates, or be uptaken by plants. If the system has reached the limits of its adsorption capacity, the contaminant removal decreases.

In all treatments in this study, the mercury concentration decreased because of the phytoremediation process, which consists of four types of plant-based technologies, namely rhizofiltration, phytostabilization, phytovolatilization, and pytoextraction [11, 27]. Among the four phytoremediation technologies, phytoextraction is the most widely used method for extracting heavy metal soil pollutants [7].

The mercury removal efficiency was experienced by all reactors from the beginning to the end of observation (Figure 4). In the 143 mg/L treatment with 100% contaminated soil, the mercury removal efficiency decreased to 52.45% on day 14 and to 39.16% on day 28. In the 238 mg/kg treatment, the mercury removal efficiency was 57.14% on day 14, reaching 65.55% on day 28. A similar trend occurred in the 143 mg/kg treatment with 80% contaminated soil and 20% compost, where the mercury removal efficiency was 48.95% on day 14 and 53.15% on day 28. In the 238 mg/kg treatment, the mercury removal efficiency was 61.76% on day 14 and 66.81% on day 28. The addition of compost to the growth media helped to increase plant growth and provided the nutrients needed for growing.

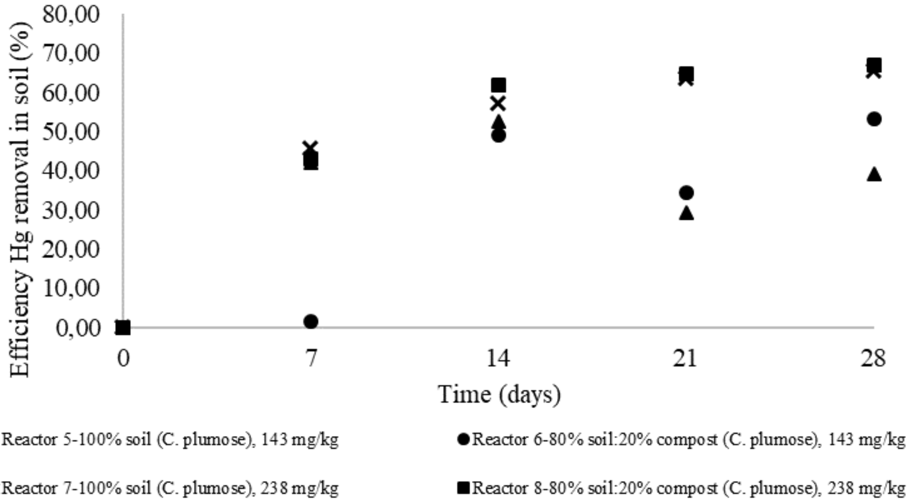

Figure 4 The mercury concentration reduction efficiency in growth media using *Celosia plumose.*

3.5 Comparison of the Efficiency Decreased Mercury Concentration in Growth Medium using *Sansevieria trifasciata* and *Celosia plumosa*

The mercury concentration in the 100% contaminated growth media decreased both with *Sansevieria trifasciata* and *Celosia plumosa* (Figure 5). The average mercury concentration reduction using *Sansevieria trifasciata* was higher on the same day of observation compared to *Celosia plumosa*, both at 143 mg/kg and 238 mg/kg. The mercury removal efficiency using *Sansevieria trifasciata* in the treatments with 143 mg/kg and 238 mg/kg on day 28 was 50.35% and 74.79%, respectively. *Celosia plumosa* could reduce the mercury concentration in the 143 mg/kg and 238 mg/kg treatments on day 28 with 39.16% and 65.55%, respectively. It can be seen that *Sansevieria trifasciata* was more tolerant to mercury and could absorb more mercury than *Celosia plumosa*. *Celosia plumosa* cannot absorb pollutant loads at high concentrations [13].

This may be because each plant has a different type of tissue so the ability and tolerance level of absorption is also different and the amount of absorbed mercury concentration also varies [25]. Yusuf, *et al.* [24] have suggested that *Sansevieria trifasciata* is stronger and can remediate soil polluted by metals. *Sansevieria trifasciata* has active compounds of pregnane glycosides. These active compounds can convert pollutants into several organic acid compounds and amino acid compounds, giving it the strength and ability to absorb heavy metals [22].

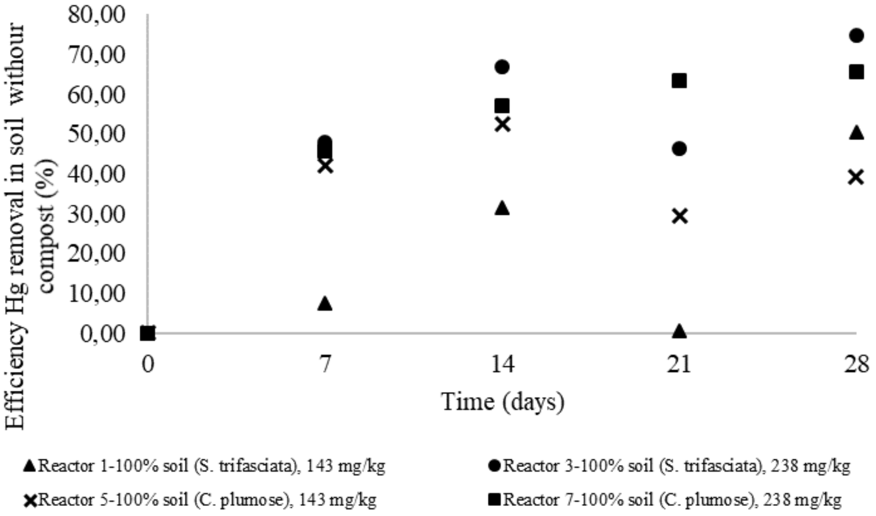

Figure 5 Comparison of the mercury removal efficiency in growth medium using *Sansevieria trifasciata* and *Celosia plumosa* without compost.

A similar trend of mercury concentration reduction also occurred in the growth media consisting of 80% contaminated soil and 20% compost (Figure 6). The mercury removal efficiency using *Sansevieria trifasciata* in the 143 mg/kg and 238 mg/kg treatments was 54.55% and 75.68% on day 28, respectively. For *Celosia plumosa*, the mercury removal efficiency levels in the 143 mg/kg and 238 mg/kg treatments on day 28 was 53.15% and 66.81%, respectively.

Growth media added with compost can absorb more mercury than growth media without compost addition because the compost increases the number of soil microorganisms that help the plants to absorb nutrients. The plants can flourish and have the power to absorb heavy metals [26]. For both types of plants, the highest absorption was found with addition of 238 mg/kg compost, because the higher the concentration, the higher the absorption [25].

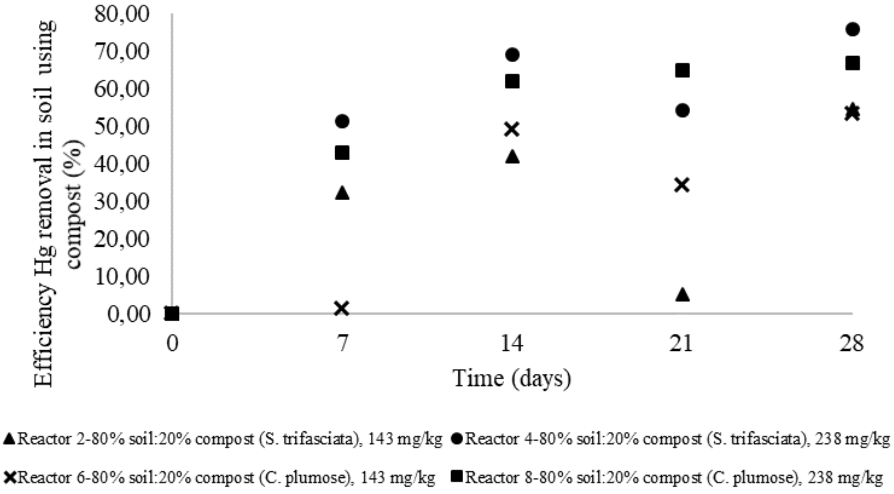

Figure 6 Comparison of the mercury removal efficiency in growth medium using *Sansevieria trifasciata* and *Celosia plumosa* with compost.

One advantage of phytoremediation is the generation of recyclable metal-rich plant residue [28]. Phytoremediation could be a viable option to decontaminate soils polluted by heavy metals, particularly when the biomass produced during the phytoremediation process can be economically valorized in the form of bioenergy. The use of metal-accumulating bioenergy crops may be suitable for this purpose. If soil contaminated by heavy metals is phytoremediated with plant oil crops, biodiesel production from the resulting plant oil could be a viable option to generate bioenergy [29].

In large-scale applications, the potential energy stored can be utilized to generate thermal energy [28]. The success of the phytoextraction technique depends on the identification of suitable plant species that can hyperaccumulate heavy metals and produce large amounts of biomass using established crop production and management practices [7].

4 Conclusions

The reactor with growth media consisting of 80% contaminated soil and 20% compost with *Sansevieria trifasciata* and *Celosia plumosa* had 75.63% and 66.81% Hg removal efficiency with the final Hg concentration at 58 mg/kg and 79 mg/kg, respectively. The reactor with growth media consisting of 100% contaminated soil with *Sansevieria trifasciata* and *Celosia plumosa* had 74.79% and 66.55% Hg removal efficiency with the final Hg concentration at 60 mg/kg and 82 mg/kg, respectively.

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