

The efficiency of pistacia atlantica extract as a natural coagulant aid on arsenic removal from aquatic environments

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Journal of Research & Health
Social Development & Health Promotion
Research Center
Vol. 5, No.1, Spring 2015
Pages: 42-49
Original Article

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Received: 29 Oct 2013

Accepted: 26 Apr 2014

How to cite this article: Bazrafshan E, Mostafapour FK, Ahmadabadi M, Afsharnia M. The efficiency of pistacia atlantica extract as a natural coagulant aid on arsenic removal from aquatic environments. *J Research Health* 2015; 5(1): 42-49

Abstract

Currently, there are millions of people around the world who are at serious risk of arsenic poisoning, in that the number of individuals exposed to arsenic in Asia is totally more than 100 million people. In this study, the ability of *Pistacia atlantica* extract was investigated as a natural coagulant aid for the arsenic removal from aqueous solutions. In this study, the effect of important utilization variables was studied such as ferric chloride coagulant dose (3-35 mg/L), *Pistacia atlantica* extract dose as the coagulant aid (0.5-5 ml/L), the initial concentration of arsenic (10-1000 µg/L), the initial turbidity (10-200 NTU) and the initial pH of the solution (2-12). The results showed that the optimum pH for maximum arsenic removal by ferric chloride is 6 and the highest removal rate for the initial concentrations of arsenic 100 µg/l was 94% using 5 mg/L ferric chloride and 0.5 ml/L *Pistacia atlantica* extract coagulant aid. The results suggest that using 0.5 ml/L coagulant aid will reduce 50% of iron chloride coagulant consumption, which is economically significant, and will also reduce sludge production and its relevant costs. However, based on the results, *Pistacia atlantica* extract is an effective coagulant aid in combination with ferric chloride for the arsenic removal from aqueous environments.

Keywords: Arsenic, *Atlantica*, Coagulant, Natural, *Pistacia*

Introduction

According to published reports, millions of people are at serious risk of arsenic poisoning, in that in many parts of the world arsenic concentration in groundwater and surface water is higher than national and international drinking water standards [1,2]. Arsenic contamination of water sources has been reported in many countries including Argentina, Bangladesh, India, Mexico, Thailand, Taiwan, Nepal, Vietnam, China, Ghana, Hungary, and even U.S. and U.K. [3,4]. It is estimated that a total of more than 100 million people are

exposed to arsenic in Asia (Bangladesh, India, China and Taiwan) [5]. In Iran some cases of contamination have been reported in Khorasan and Kurdistan provinces. According to study of Mosaferi et al., the concentration of arsenic in drinking water in 10 villages in Kurdistan exceeds permissible concentration and limits of changes have been reported from 10-500 µg/L [4]. Arsenic compounds in groundwater, as an important source of drinking water, is a serious health and environmental issue [6] and has been known as a major contaminant in drinking water, especially in the South

Asian regions. In these areas, millions of people are at risk of arsenic-related diseases [2,7]. Arsenic contamination of groundwater occurs due to a natural process such as erosion of mineral containing arsenic and also human activities such as uncontrolled industrial output from mining and metallurgical industries, insecticides, and organoarsenical pesticides [6]. According to International Association on Research Cancer, inorganic arsenic compounds are placed in group 1 (carcinogenic to humans) [7,8]. The toxicity of arsenic depends on the chemical and physical properties of compounds, the entrance to the body, the dose, duration of exposure and its value in the diet and age. Acute poisoning with arsenic affects the central nervous system and leads to coma. Chronic poisoning causes a general weakness in the muscles, loss of appetite, nausea, inflammation of the mucous membranes of the eyes, nose, throat and skin lesions. Neural presentations and even malignant tumors in vital organs caused by arsenic have been found [8,9]. Generally, exposure to arsenic through drinking water causes various types of skin lesions, skin and liver cancer, cardiovascular and neurological disorders and it also affects the hematopoietic system [1,8,10]. Water pollution with arsenic has made water and health officials and relevant authorities provide strict standards for arsenic in drinking water [3], so that since 2006 WHO and USEPA have lowered the maximum contaminant level (MCL) from 50 to 10 $\mu\text{g/l}$ [6,8]. According to the standard of drinking water in Iran, the maximum contaminant level of arsenic is 10 $\mu\text{g/l}$ [9]. Strict rules for arsenic in drinking water have led to an important challenge in conventional water treatment technologies [8], so that it has made water authorities determine, identify and use appropriate and applicable techniques at reasonable prices [3]. U.S. EPA has introduced seven technologies as the best available technologies (BAT) for the arsenic removal including the process of ion exchange, activated alumina, reverse osmosis, modified coagulation/filtration, lime-modified lightweight, reverse electrodialysis,

oxidation/filtration [6], among these methods, coagulation process has the highest application for the removal of high concentrations of arsenic from water due to low cost, high efficiency and being widely used in developing countries [10]. In addition, the coagulation process can effectively remove arsenic along with dissolved and suspended compounds in water especially turbidity, color, phosphate and fluoride [1]. In Taiwan, the coagulation process using iron ions for purifying well water containing high concentrations of arsenic dates back to before 1960. According to reports, in the conventional treatment with alum and ferric ions as a coagulant, arsenic is removed with high efficiency [10]. Using these coagulants alone will lead to large quantities of sludge production, so that they cause concerns about environmental effects when entering the environment in large volumes as a secondary sludge, therefore, coagulant aids are required [11]. In recent years, a particular interest has emerged for using natural coagulants in water treatment in developing countries for their advantages such as biodegradability, less sludge production and minimum environmental and health risks [12]. *Pistacia atlantica* is one of the wild pistachio species that spreads from the Canary Islands and the Mediterranean countries to Asia Minor, Syria, the Caucasus, Iran, Afghanistan and Pakistan. *Pistacia atlantica* in Iran is seen massively between the provinces of Fars and Kurdistan and sporadically in other parts of the country. In Iran, *Pistacia atlantica* cover more than one million two hundred thousand acres of forests. *Pistacia atlantica* extract is one of the natural anionic coagulants and its kernel contains more than 65% of *Pistacia atlantica* seed weight whose oil value is 60% (with oleic acid 50.4%, linoleic acid, 30.8%, and palmitic acid 12.2%). Considering that Iran has huge resources of *Pistacia atlantica* and also due to many advantages such as ease of access, low cost, and being natural (regarding the reduction of environmental pollutions), this study was designed and implemented to

evaluate *Pistacia atlantica* extract efficiency as a natural coagulant aid with chemical coagulant ferric chloride for the arsenic removal.

Method

This study pilot-scale experimental study was conducted in a closed system. In all stages of experiments, double-distilled water was used to accurately control the situations. To study the effect of variables of coagulant dose (3-35 mg/L), coagulant-aid dose (0.5-5 ml/L), the initial concentration of arsenic (10-1000 µg/L), the initial turbidity (10-200 NTU), and pH of solution (2-12), experiments were conducted by a 2-liter jar in a jar test device programmed for 2 minutes rapid mixing at 200 rpm, 15 minutes slow mixing at 40 rpm and 30 minutes sedimentation [13].

Light kaolin suspension was used according to Smith and Cohen method to create turbidity in participants [14]. To prepare the coagulant aid, *Pistacia atlantica* seeds were washed with tap water to remove dust and then rinsed with distilled water. Then the seeds were dried in an oven at 35°C for 5 hours. In the next stage, the seeds were peeled manually and then were milled and sieved with a screen 0.4 mm and the part with the size of less than 0.4 was used in experiments. Five grams of the resulting powder was dissolved in 1000 ml of 0.5 molar NaCl and then the resulting suspension was stirred for 10 min and the resulting solution

was filtered with a 0.45 micron filter. This solution was used as a coagulant aid. The above solution was prepared freshly and kept in a cool place with a maximum temperature of 4°C to avoid changes in pH and viscosity [14,15].

Stock arsenic solution (1000 mg/L) was prepared by $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ and kept in the refrigerator at 4°C [6]. To provide 100 mg/L alkalinity in carbonate, NaHCO_3 was used [16,17] and to adjust pH of participants, 1 molar NaOH and HCl solutions were used. After conducting experiments, sampling was performed from the depth of 2 cm [18] and arsenic concentration in the participants was measured according to standard method sodium diethyldithiocarbamate silver and by a spectrophotometer UV-VI (PG, Instruments Ltd) with wavelength 520 nm [19].

Results

In Figure 1, the effect of PH on arsenic removal efficiency is shown in constant conditions of ferric chloride dose 10 mg/l and an initial arsenic concentration 100 µg/L. According to figure 1, the greatest amount of arsenic removal was obtained 90.89% at pH=6, while at higher and lower pHs, the removal efficiency decreased significantly. The concentration of arsenic remaining in solution reached to approximately 9.1 µg/L. Thus, pH=6 was used for other experiments.

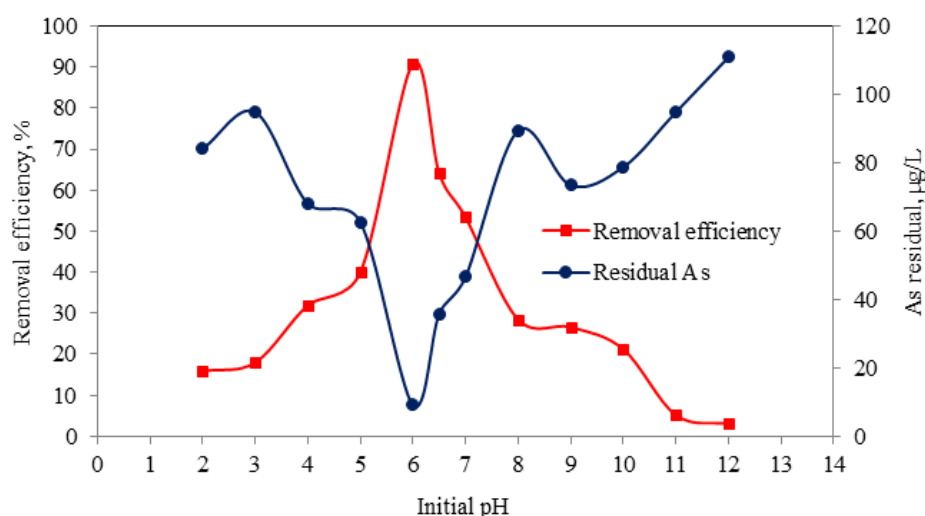


Figure 1 Effect of initial pH on As removal efficiency (FeCl_3 dose = 10 mg/L, As concentration = 100 µg/L)

In Figure 2, the effect of different doses of ferric chloride coagulant for arsenic removal is studied under optimal conditions pH=6 and initial arsenic concentration 100 µg/L. According to figure 2, by increasing ferric chloride coagulant dose from 3 to 10 mg/l, removal efficiency was significantly increased

(increased from 48.03% to 92%) but with further increase in coagulant dose the removal efficiency showed no significant increase, so that by increasing coagulant dose from 10 to 35 mg/l the removal efficiency changed from 92% to 96%.

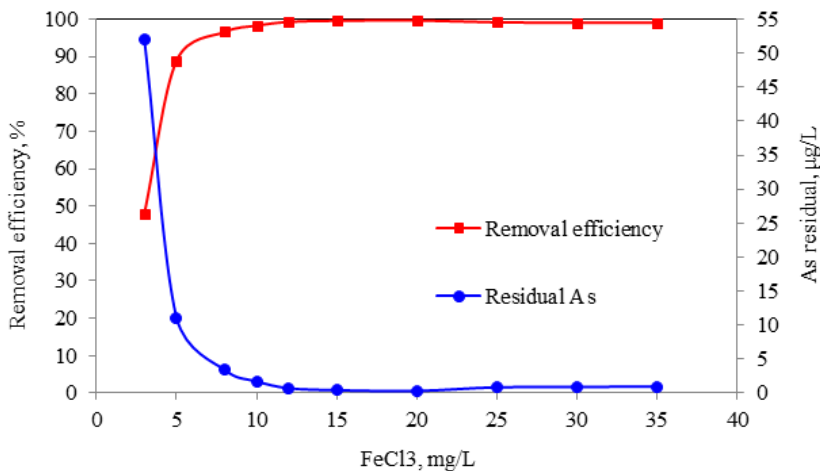


Figure 2 Effect of coagulant dose (3-35 mg/L) on As removal efficiency (As concentration= 100 µg/L, pH=6)

In Figure 3, the effect of initial arsenic concentration on the removal efficiency in optimal conditions pH=6 and ferric chloride coagulant dose 10 mg/L is studied. According to figure 3, arsenic removal efficiency is reduced

by increasing its initial concentration, so that the removal efficiency changed from 100% for the arsenic concentration 40 µg/L to 28% for the arsenic concentration 1000 µg/L.

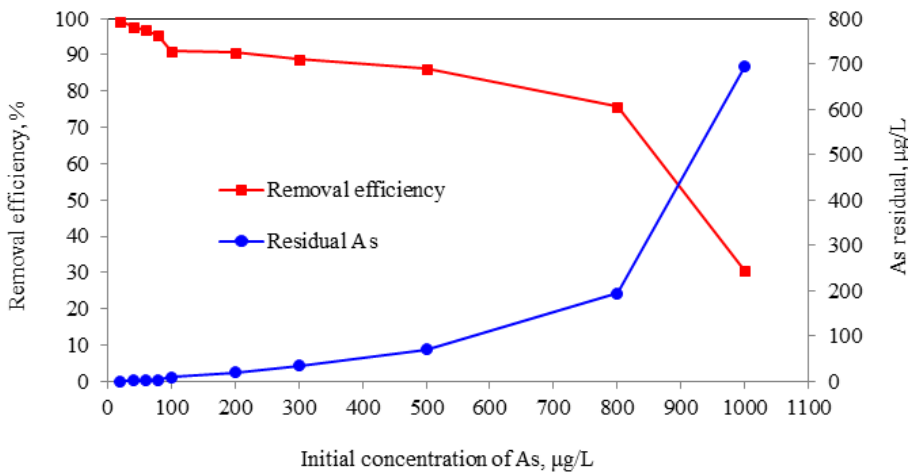


Figure 3 Effect of initial concentration of As on removal efficiency (pH=6, coagulant dose of FeCl3= 10 mg/L)

To examine the effect of coagulant aid in the arsenic removal, ferric chloride doses less than optimal amounts were used in combination with a coagulant aid. In Figure 4, the effect of Pistacia atlantica extract coagulant aid in the arsenic removal in conditions of pH=6, ferric chloride 3 and 5 mg/l and arsenic 100 µg/l was studied. As observed,

the highest removal efficiency in coagulant aid dose 1 ml/l was 65.53%, while without using coagulant aid the removal efficiency was only 48.03%. furthermore, the maximum removal efficiency was obtained 93% in coagulant aid dose 0.5 ml/L, while without using coagulant aid the removal efficiency was 75%.

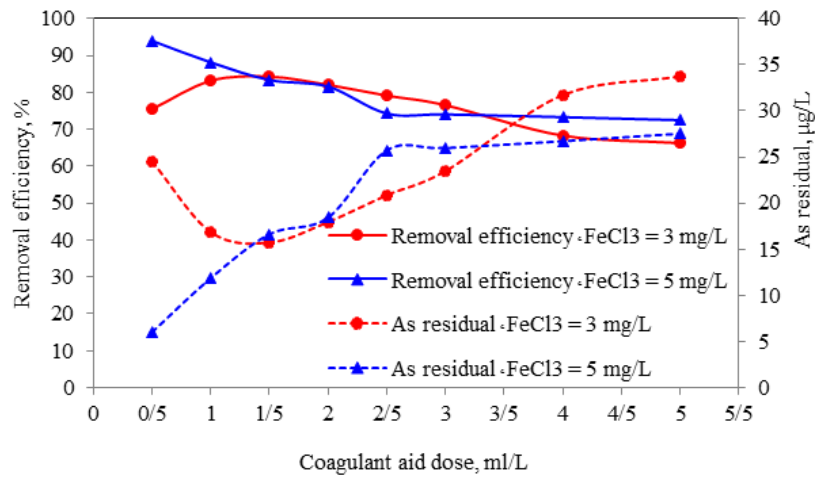


Figure 4 Effect of coagulant aid dose on As removal efficiency (pH=6, FeCl3= 3 and 5 mg/L)

In Figure 5, the effect of turbidity on arsenic removal under optimal conditions pH=6, concentration of ferric chloride 5 mg/l, arsenic concentration 100 µg/l and the concentration of coagulant aid 0.5 ml/l are shown. According to figure 5, by increasing

turbidity to 20 NTU the removal efficiency was increased to 97% and with further increase in turbidity from 20 to 200 NTU the arsenic removal efficiency decreased from 97% to 63%.

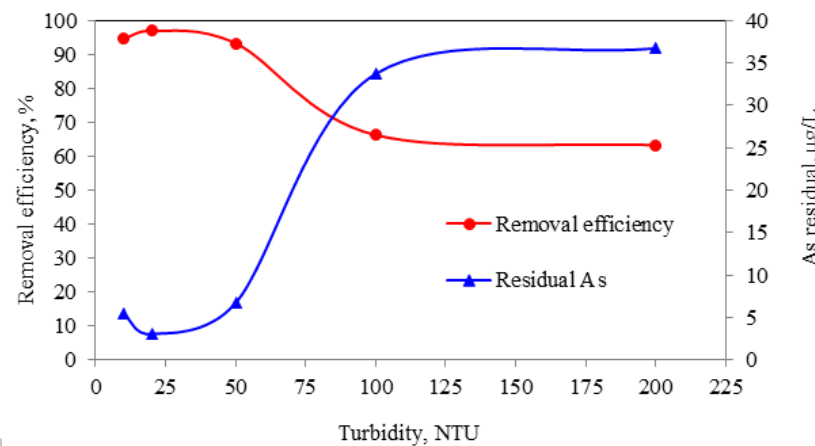


Figure 5 Effect of initial turbidity on As removal efficiency (As concentration= 100 µg/L, pH=6, FeCl3 = 5 mg/L, Coagulant aid dose= 0.5 ml/L)

Discussion

The results of this study showed that the optimum pH for the maximum arsenic removal by ferric chloride coagulant was 6 and the highest removal rate for the initial concentration of arsenic 100 µg/l was 94% by using 5 mg/L ferric chloride and 0.5 ml/L Pistacia atlantica extract coagulant aid. The findings of this study also suggest that using 0.5 ml/L Pistacia atlantica extract coagulant aid can reduce 50% of iron chloride coagulant consumption during the removal process. In the following, the effect of important utilization variables is discussed on the process of arsenic removal from aqueous environments.

pH of a solution is considered one of the most important parameters affecting coagulation process and pollutants removal from aqueous solutions. To determine the optimum pH for the arsenic removal by Pistacia atlantica extract as a natural coagulant, the arsenic removal was investigated as a function of hydrogen ion concentration. In Figure 1, the effect of pH on arsenic removal efficiency is examined. According to the figure, the greatest amount of arsenic removal at pH=6 was obtained 90.89%, whereas at higher and lower pHs, the removal efficiency was significantly reduced. Since the absorption

of the main mechanism of arsenic removal (As (V)) is by coagulation with ferric chloride, and at acidic pHs ($\text{pH} < 4$) arsenic is also converted into uncharged and nonabsorbable species [20] and on the other hand, in alkaline pHs ($\text{pH} = 7-11.5$) arsenic is in the form of HAsO_4^{2-} and in acidic pHs ($\text{pH} < 7$) is in the form of H_2AsO_4^- and since arsenic in the form of H_2AsO_4^- is better removed than arsenic in the form of HAsO_4^{2-} , the removal efficiency is reduced by increasing and decreasing pH [21]. However, by increasing pH, we approach to the isoelectric point ($\text{pH} = 7.9$) of ferric hydroxide, during which the surface potential of ferric hydroxide is reduced [22]; thus, by increasing pH the active sites with positive charge are reduced on the surface of ferric hydroxide and lead to the reduction of arsenic removal efficiency in the alkaline pHs [7]. The results of this study were consistent with data obtained from Pakzadeh studies (pH between 4.5 and 9) [24]. Also, in Akshmanan et al.'s study titled as 'a comparative study of arsenic removal by iron ions by chemical and electrical coagulation', the results showed that in pH range of 6.5, 7.5 and 8.5, the highest arsenic removal efficiency in chemical coagulation was obtained by ferric chloride at $\text{pH} = 6.5$ [25]. Also based on Baskan et al.'s study, the best pH (4 to 9) for arsenate removal by ferric chloride was 7.5 [25]. According to the results of some studies, the optimal pH range is 6 to 8 [23,26,27].

Based on the results presented in Figure 2, it is observed that the increase of coagulant dose caused a significant increase in arsenic removal. Adding coagulant will facilitate arsenate removal from aqueous environment by converting the species of As (V) solution into insoluble products by providing more hydroxide surfaces to absorb As (V). Accordingly, the results of this study are consistent with the results of Pakzadeh et al. (initial concentration of arsenate 9.7 mg/L and dose of ferric chloride in Fe, 10 to 80, that the removal efficiency was increased by increasing of coagulant dose) [20]. The results are also consistent with the results of Lakshmanan et al. [24] and Baskan et al. [25].

According to Figure 3, the arsenic removal efficiency is decreased by increasing its initial concentration, since the absorption process of the main mechanism of arsenic removal is performed by coagulation with ferric chloride. With increasing the initial concentration of arsenic and constant coagulant concentration, the number of available sites is not sufficient to absorb all arsenic ions, so the removal efficiency is reduced, so that the removal efficiency changed from about 99% for arsenic concentration of 20 $\mu\text{g/L}$ to 30.42% for arsenic concentration of 1000 $\mu\text{g/L}$. The results of this study are consistent with the studies of Pakzadeh [20], Baskan [25], Wu [28] and Song et al. [10].

According to Figure 4, by increasing coagulant dose of *Pistacia atlantica* extract, arsenic removal efficiency is increased up to a maximum value and more increase in coagulant aid led to the reduced removal efficiency. The use of lower doses of coagulant aid means that there are not enough chain molecules for bridging between micro flocks, while higher doses may increase the zeta potential towards more negative values and this leads to arsenic removal ions with negative charge, so only appropriate and adequate doses are effective on coagulation process performance [29]. As observed, utilizing 0.5 ml/L coagulant aid with 5 mg/L ferric chloride in initial arsenic removal 100 $\mu\text{g/L}$, leads to 94% reduction of the remaining arsenic, however, to achieve this removal efficiency by using ferric chloride without coagulant aid, 10 mg/L ferric chloride should be used. The results suggest that using 0.5 ml/L coagulant aid will reduce iron chloride coagulant consumption by about 50%, which economically significant, and will also reduce sludge production and costs related to it.

Studying the effect of different initial turbidities on the arsenic removal in the initial concentration of arsenic 100 $\mu\text{g/L}$ (Figure 5) indicates that adding turbidity to the participant can lead to a slight increase in arsenic removal compared to using coagulant individually, because turbidity can provide

conditions for more collision of particles with each other and thus increase in removal efficiency. However, adding more turbidity was associated with reduced arsenic removal, probably because of consuming the majority of the coagulant to remove turbidity. In other words, the absence of turbidity and thus the absence of sites that act as the primary cores for creating clots are evidence of poor formation of large clots and the lack of sedimentation during 30 minutes [30], so turbidity leads to some extent to increased removal efficiency. The results of this study are consistent with studies of Meng et al., results of Bazrafshan et al. on arsenic removal by using *Moringa Peregrina* seed extract as a natural coagulant and also the study of Antov et al. on the removal of turbidity from aquatic environments by using proteins in bean seeds as natural coagulant [22,31,32]. Finally, according to results, given the ease of preparation and high efficiency in removing arsenic, the use of this natural coagulant can be considered by the relevant authorities, although it is only present in some parts of the country.

Conclusion

The results of experiments suggest that *Pistacia atlantica* extract is an efficient and reliable natural coagulant aid in combination with ferric chloride for removing arsenic from the water, so that the percentage of arsenic removal by adding *Pistacia atlantica* extract coagulant aid shows an uptrend due to surface absorption and interparticle bridging. According to results, it seems that using *Pistacia atlantica* extract coagulant has reduced 50 percent of ferric chloride which reduces procurement costs of coagulant and will lead to decreased residual iron in water and minimize its health complications. Furthermore, less sludge production and its removal costs will be also reduced. The results also show that *Pistacia atlantica* extract coagulant aid with ferric chloride can provide standard arsenic in drinking water at initial concentrations of 100 µg/l.

Acknowledgments

The authors would like to express their

appreciation to Department of Environmental Health Engineering and Health School of Zahedan University of Medical Sciences for the financial support of this study (Project NO: 5731).

Contributions

Study design: EB,FKM

Data collection and analysis: EA

Manuscript preparation: EB, MA

Conflict of interest

"The authors declare that they have no competing interests."

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