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To cite this article: Xuejiao Qi *et al* 2017 *IOP Conf. Ser.: Earth Environ. Sci.* **81** 012018

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Study on natural soil coagulants for point-of-use drinking water treatment

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Abstract: Natural soil coagulants were used for domestic drinking water treatment in Ethiopia because of lacking drinking water supply. In this study, two soils were collected from Ethiopia and jar test was used to investigate the optimum working conditions, performance and safety of them. The turbidity removal efficiencies of soil A and soil B are 74.5% and 91.1% (initial turbidity=400 NTU), respectively. The pH of purified water can meet drinking water standards of Ethiopia (pH=6.5-8.5). Both soils contribute to remove TOC when the initial concentration of it is high. A small amount (0.5 mg. L⁻¹ or 1 mg. L⁻¹) of poly-aluminum chloride can improve the performance of soil coagulants for the removal of turbidity. The application of soil coagulants didn't introduce coliforms, although the total bacteria number of treated water increased to 40 CFU (soil A) and 81 CFU (soil B), still meeting the drinking water quality standards (≤ 100 CFU).

1. Introduction

The population growth and lack of infrastructure had made it a serious problem to get safe drinking water for people in developing countries, such as Ethiopia [3]. High turbidity, which decreases the quality of drinking water, is one of the major problems of sources of drinking water in Africa. For instance, in Nairobi, the turbidity of raw water can be 3 NTU in the dry season, and it may reach to 5000 NTU during the rainy season [17, 18]. It is reported that the turbidity of Blue Nile and River Nile may reach 7,275 NTU and 6,575 NTU, respectively [11].

Conventional chemical-based coagulants, such as alum, ferric chloride and poly-aluminum chloride (PAC) are extensively used to remove turbidity from water [6]. However, the cost of importing these kinds of chemical coagulants may be too high for many developing countries. Natural local coagulants may be one of the best choices due to their low price and environment-friendly characteristics. Some natural coagulants, such as *Moringa oleifera* seeds [3], alginate [15], chitosan [21], *strychnos potatorum* and *Pistacia atlantica* seed extract [2], were used to treat drinking water or industrial wastewater.

In Africa, local soils are a kind of natural coagulant which have been used for point-of-use drinking water treatment [12, 14] for hundreds of years [5] because of the lack of centralized drinking water treatment and the high turbidity of source water. People in some rural areas can't afford to use synthetic chemical coagulants to purify water. However, the use of natural soil coagulants was seldom put into research. It was reported that lots of Ethiopians use local soils to purify drinking water



according to Central Statistical Agency (CSA) [5]. Eyasu Getahun mentioned that people living in Boke-Tino, Ethiopia, use different kinds of soils to treat drinking water now [5].

Besides low cost and sufficient quantity, clay minerals have high cationic exchangeable capacity, and they had already been used as coagulants to treat water [7, 8]. However, there is a lack of systematic and detailed study about the application of natural soils. The soil coagulants studied in this paper are used for point-of-use drinking water treatment [12, 14]. Therefore, the purpose of this research is to study the optimal conditions of these soils for domestic water treatment and to investigate possible mechanism of coagulation. We have studied the optimum dosage and settling time, investigated effects of pH and total organic carbon (TOC), and explored the combination use of soils and PAC, detected total coliform groups and total bacterial colony number after coagulation by soils.

2. Materials and Methods

2.1 Collection and preparation of natural soil coagulant

Both soil A and soil B were collected from Boke-Tino, located at the middle of Ethiopia. Soils were dried under 50°C first, and then were ground to pass through a 100 mesh sieve. The basic composition of soil A and soil B are shown in table 1.

Table 1 Basic composition of soil A and soil B (g·kg⁻¹)

Sample	TOC	TN	Al	Ca	Fe	K	Mg	Mn	Pb	Zn	Cd	Cr	Cu
Soil A	10.13	0.20	12.34	156.69	11.22	2.97	0.42	0.22	0.10	6.01	0.64	6.26	6.10
Soil B	13.43	0.90	4.76	147.74	6.93	1.23	0.08	0.05	0.02	8.58	0.41	0.99	1.45

*TOC: Total organic carbon; TN: Total nitrogen

2.2 Preparation of synthetic water, PAC and humic acid solution

Synthetic water of different turbidity was prepared by adding kaolin into tap water. The quality of tap water was shown in table 2. 10 g·L⁻¹ kaolin stock suspension solution was made first, and other desired concentrations of turbid water were prepared by diluting suspension of stock solution using tap water.

Table 2 The quality of tap water

Turbidity(NTU)	pH	TDS(mg/L)	EC(μs/cm)
0.25	7.5	188.1	378

*TDS: Total dissolved solids; EC: Electrical conductivity

The stock solution of PAC was made by adding 0.2 g PAC into 100 mL deionized water. During coagulation test, different dosages (0.5 mg·L⁻¹, 1 mg·L⁻¹, 3 mg·L⁻¹ and 5 mg·L⁻¹, respectively) of PAC solution was added into 200 mL suspensions of kaolin solution.

The humic acid (HA) was purchased from Sigma-Aldrich, and the basic features have been characterized in others' work [20]. In addition, our previous work has presented the features of this HA [10]. The stock solution of HA was prepared by adding 0.16 g HA into 100 mL deionized water. Then different dosages of HA stock solution was added into 200 mL turbid water just before coagulation test. The concentrations of TOC were determined to be 2.94 mg·L⁻¹, 4.66 mg·L⁻¹ and 11.37 mg·L⁻¹, respectively.

2.3 Measurements of turbidity, TOC, electrical conductivity, zeta potential and particle size

A 2100Q Portable Turbidimeter (HACH, USA) was used to measure the turbidity. The removal efficiency of turbidity was calculated as below:

$$\text{Removal efficiency} = \frac{\text{initial turbidity} - \text{final turbidity}}{\text{initial turbidity}} \times 100\%$$

A Shimadzu TOC V-CPN (Shimadzu, Japan) was used to measure the TOC of synthetic water. The electrical conductivity was measured by a conductivity meter DDSJ-318 (Inesa, China). The zeta

potential of water samples was measured by a Malvern Zetasizer (Nano ZS 90, UK). The particle size of soils was measured by Laser Scattering Particle Size Distribution Analyzer LA-960 (Horiba, Japan).

2.4 Coagulation test

Jar test was conducted to evaluate coagulation process of soils under different conditions. First, 200 mL synthetic water was added into 250 mL beaker, and then 0.1 mol/L HCl and 0.1 mol/L NaOH were used to adjust pH. The desired concentration of TOC was obtained by adding HA solution into turbid water. Different dosages of soil were added into each beaker with a rapid stirring of 300 r/min for 5 minutes, followed by a slower stirring of 100 r/min for 15 minutes. Finally, the residual turbidity was measured after settling for a certain time (ranging from 30 minutes to 8 hours) according to different purposes.

In order to investigate the influences of ionic strength, NaCl was used to adjust the electrical conductivity of water. The initial EC is 2 mS/cm, 6 mS/cm and 10 mS/cm, respectively, and the coagulation test was repeated.

2.5 Detection of total coliform groups and bacterial colony number

To investigate the microorganism risk of soils, we measured the coliforms and bacterial colony number in the treated water. Firstly, tap water was boiled for 10 minutes to avoid the influence of residual chlorine and microorganism. Secondly, jar test was performed according to the above-mentioned steps (Coagulation test). Finally, four kinds of water sample were prepared including distilled water, 400 NTU kaolin suspension, water treated by soil A or soil B, respectively. In addition, all apparatus were sterilized by moist heat sterilization before using, and coagulation process was conducted near the flame of alcohol burner.

The coliform groups were tested by multiple-tube fermentation technique, and the colony form unit technique (CFU) was used to test bacterial colony number.

3. Results And Discussion

3.1 The effects of soil dosage on coagulation

Fig.1 shows the removal efficiency of turbidity under different dosages of coagulant after 2 hours settling.

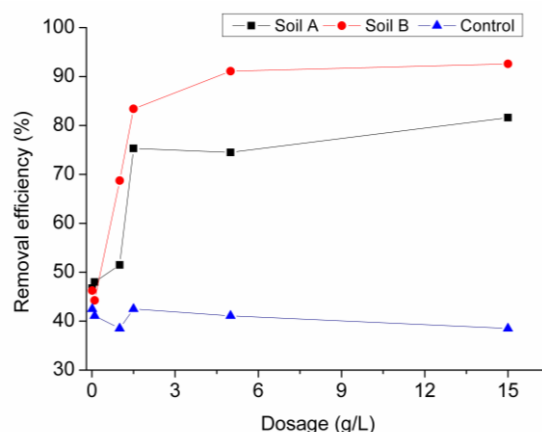


Figure 1. Turbidity removal efficiency of different dosages of soils after 2 hours settling (initial turbidity=400 NTU)

It can be found that both of the coagulants performed better with higher coagulant dose, and the turbidity removal efficiency of soil A and soil B was found to be the highest (81.6% and 92.6%, respectively) when the dosage is 15 g. L⁻¹. However, Fig. 1 shows that the removal efficiency of turbidity of soils has reached 74.5% and 91.1% (soil A and soil B, respectively) when the dosage is 5 g. L⁻¹. The cost should be considered because people need to pay for the soils. Our on-site investigation in Ethiopia shows that the dosage used there is about 5 g. L⁻¹. Therefore, 5 g. L⁻¹ was selected as a reasonable and practical dosage in this study.

3.2 The effects of initial turbidity and settling time on coagulation

In consideration of the huge variation of turbidity in raw water in Ethiopia, three kinds of water with different initial turbidities were chosen in this study, including high turbidity (745 NTU), middle turbidity (370 NTU) and low turbidity (85 NTU).

Fig.2 shows that, when low turbidity water was tested, the removal efficiency of turbidity for soil A, soil B and control (without soils) is 31.9%, 28.8% and 38.6% respectively even after 2 hours settling. Therefore, it can be concluded that soils showed little capacity to treat low turbidity water. On the contrary, the removal rate of soils for middle turbidity or high turbidity water is above 79.4%, which is higher than the control (42.0%). Therefore, soils may be appropriate in the treatment of high or middle turbidity water. In consideration of the fact that the turbidity of raw water in Ethiopia is usually close to the middle turbidity level [5], we use middle turbidity (400 NTU) as the initial turbidity in this study. According to Fig. 2, two hours is optimal settling time for both coagulants at middle and high turbidity.

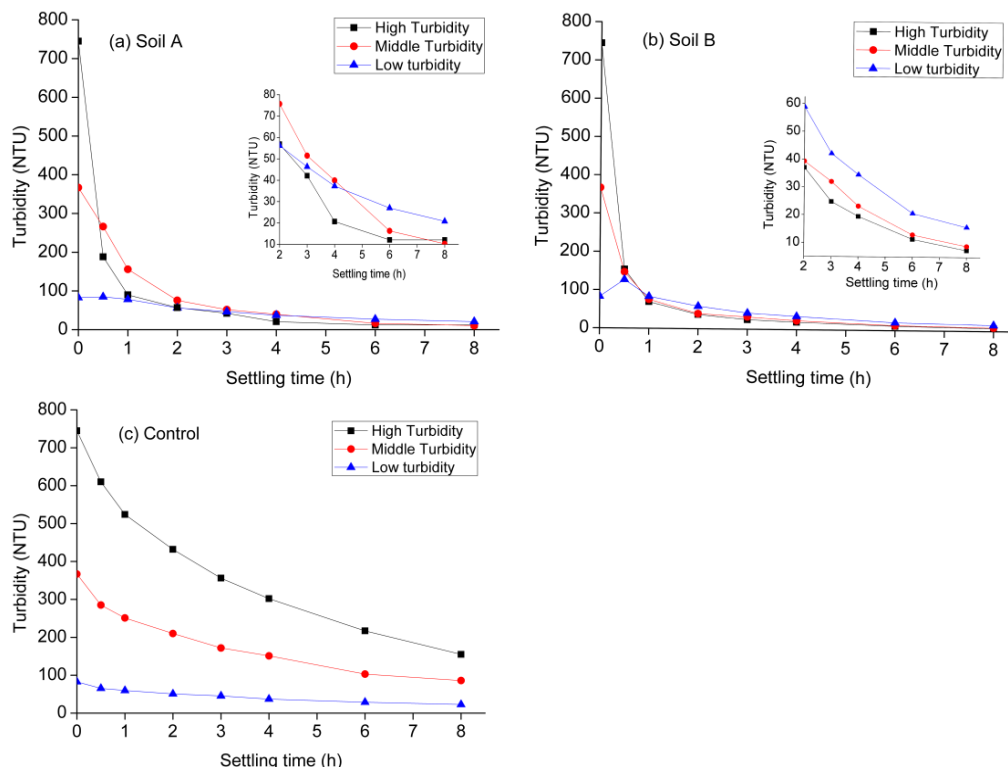


Figure 2. Final turbidity of different turbid water at different times

Fig. 2 shows that, when the initial turbidity is 370 NTU, the turbidity removal efficiency of soil B (89.3%) is much higher than that of soil A (79.4%) after 2 hours settling. As described in Fig. 3 (a), the average electrical conductivity (EC) of water treated by soil B is 2223 μ S/cm, which is higher than that by soil A (1539 μ S/cm). Fig.3 (b) demonstrates that the turbidity removal efficiencies of both soils (after 2 hours) increase with the increase of EC. And the final removal efficiencies of soil B are always higher than that of soil A because the EC of soil B is higher than that of soil A. Therefore, soil B results in higher ionic strength and contributes to the process of coagulation by condensing the electric

double layer [16].

In addition, the absolute values of zeta potentials were determined to be lower than 5 mv, which indicates that charge neutralization isn't the main mechanism of the soil coagulation [9, 19].

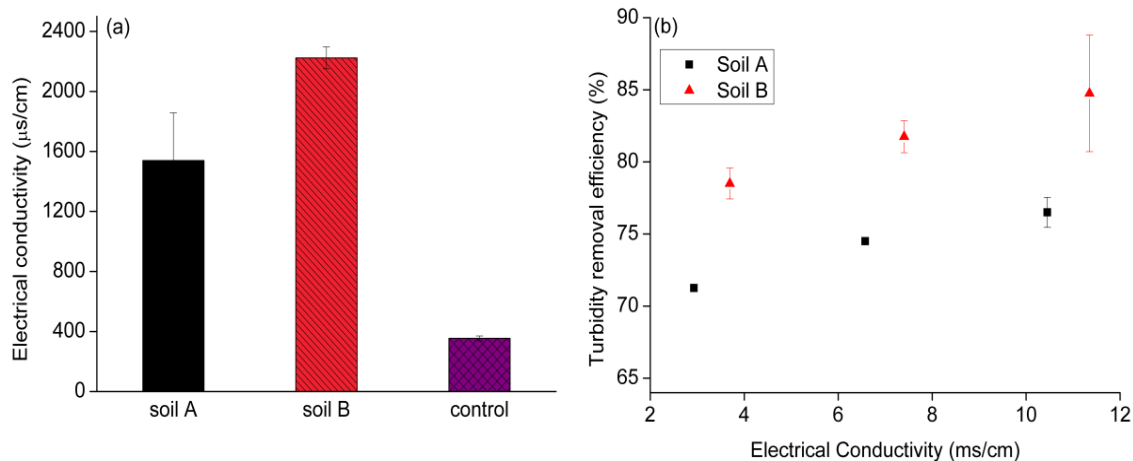


Figure 3 (a) The average electrical conductivity of different soils; Figure 3 (b) The influences of electrical conductivity on turbidity removal efficiency

3.3 The effects of pH on coagulation

Fig. 4 (a) shows that both soil A and soil B are not pH sensitive when they are used to remove turbidity, and the removal efficiency of both coagulants ranged between 85% and 93%. In contrast, the control test indicates that the removal rate is the highest at pH=7.

Fig. 4 (b) indicates that the final pH varied from 7.4 (initial pH=5) to 8.5 (initial pH=9), and this pH range meets the drinking water standards of Ethiopia ($6.5 < \text{pH} < 8.5$). Therefore, both soil A and soil B have buffering capacity to maintain a stable pH range.

Fig.4 (b) indicates two possible mechanisms why the removal efficiency of soil B is better than that of soil A. First, the final pH of water treated by soil B is closer to 7.0, where coagulants acquire the highest removal efficiency. Second, the electrical conductivity of soil B is much higher than that of soil A. Therefore, soil B may result in higher ionic strength and contribute to the coagulation process by condensing electric double layer.

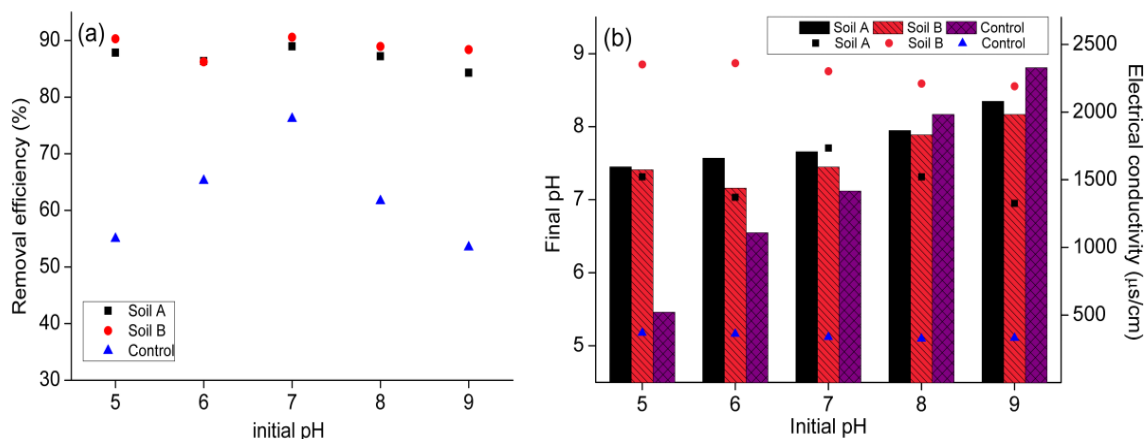


Figure 4 (a) The effects of initial pH on turbidity removal efficiency; (b) on final pH and EC after 2 h settling (In Figure 4 (b), bar graph is final pH, and dot pattern is EC)

3.4 The effects of TOC concentrations on the turbidity and TOC removal efficiency

Fig.5 (a) shows that the removal rate of turbidity increased as the concentration of TOC increased, and was higher than the control (TOC=2.94 mg. L⁻¹). HA is a kind of natural macromolecule organic matter, which may contribute to efficiency of coagulation by bridging and sweeping [1, 13, 19].

Fig. 5 (b) indicates that TOC removal efficiency of soil A is higher than that of soil B. When the concentration of TOC is 2.94 mg. L⁻¹, the TOC removal efficiency of soil B is -21.29%. The result of TOC measurement shows that the TOC of soil A is 10.13 g. kg⁻¹, and that of soil B is 13.43 g. kg⁻¹. Therefore, soil B may introduce more TOC to the water than soil A during coagulation process. And the residual TOC of water purified by both soil A and soil B is becoming similar with the increasing of TOC concentrations. In conclusion, the reasons why soil A behaves better than soil B in removing low concentration of TOC may be that soil B contains more organic matter than soil A.

It should be pointed out that these soils are not suitable for removal of organic matters. However, the removal of organic matter is sometimes of critical importance (e.g. minimizing the formation of disinfection by-products). Potential counter measures (such as activated carbon adsorption) may be applied in such situation.

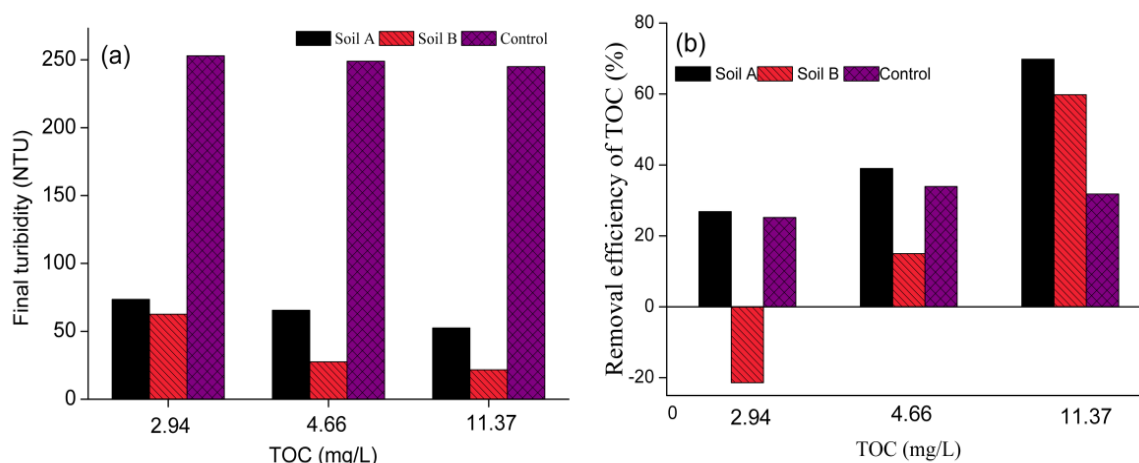


Figure 5 (a) The effects of initial concentrations of TOC on final turbidity; Figure 5 (b) The effects of initial concentrations of TOC on the removal efficiency of TOC

3.5 Combined use of soils and PAC

Fig. 6 shows the combination use of soils and PAC. Fig. 6 (a) and Fig.6 (b) showed that the final turbidity of water treated by the combination of soils and PAC is lower than the control (coagulation by PAC without soils) when the dosage of PAC is 0.5 mg. L⁻¹ or 1 mg. L⁻¹. However, Fig. 6 (c) and Fig. 6 (d) indicated that the result was opposite when the dose of PAC increased to 3 mg. L⁻¹ or 5 mg. L⁻¹. Therefore, both soil A and soil B contributed to the removal of turbidity when 0.5 mg. L⁻¹ or 1 mg. L⁻¹ PAC was used, even though the final turbidity of all of these kinds of purified water is still higher than the drinking water standard of Ethiopia (5 NTU).

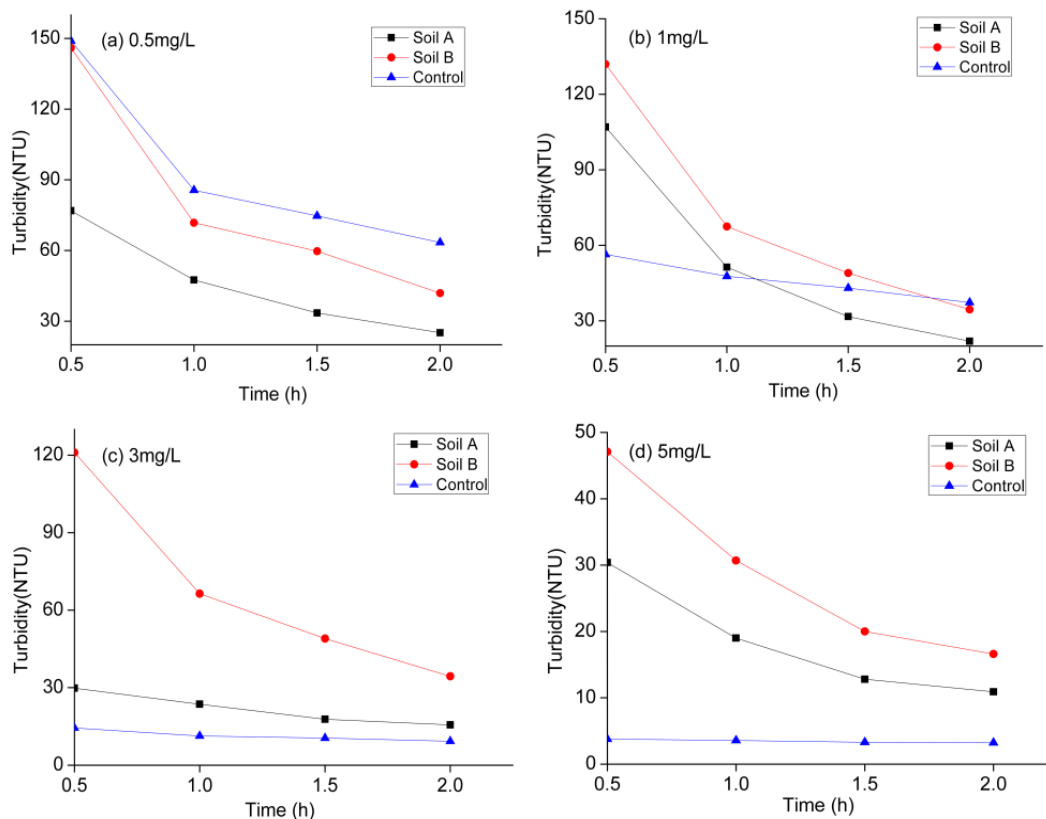


Figure 6. Change of turbidity under different dosages of PAC over time

Another interesting result is that the performance of soil A in combination with PAC is better than that of soil B in combination with PAC. One possible reason may be that the particle size of soil B is much smaller than that of soil A. The average particle size of soil A is $106.35\ \mu\text{m}$, and that of soil B is $54.41\ \mu\text{m}$. The medium diameter of soil A and soil B is $98.99\ \mu\text{m}$ and $38.88\ \mu\text{m}$, respectively. In addition, soil B is powdered, and soil A is sand-like particles. Therefore, it is easier for soil B to be suspended in water and to result in higher turbidity.

Fig. 7 (a) indicates that the pH of the water before and after adding PAC is stable (about 8.0). Fig. 7 (b) shows that the electrical conductivity of soil B is much higher than that of soil A and the control, which indicates that condensing the double layer does not play the main role during the coagulation-flocculation process. Therefore, the combined use of soil A and $0.5\ \text{mg}\cdot\text{L}^{-1}$ or $1\ \text{mg}\cdot\text{L}^{-1}$ PAC could improve turbidity removal efficiency, and the electrical conductivity of treated water meets the drinking water standards of World Health Organization (WHO) ($<2000\ \mu\text{S}/\text{cm}$). In addition, the absolute value of zeta potential is measured to be near zero ($0.0875\sim 5.280\ \text{mV}$), which indicated that charge neutralization doesn't play the main role.

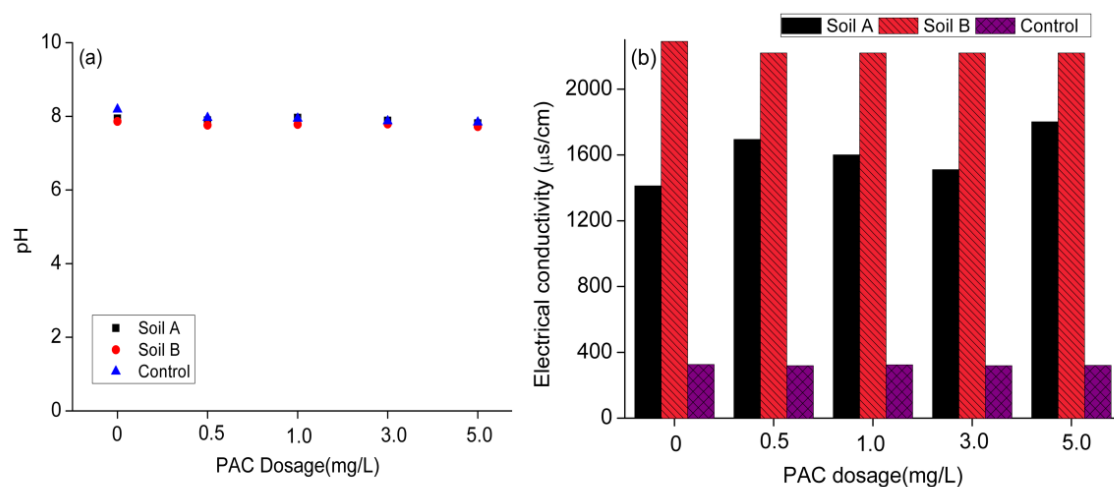


Figure 7 (a) The effects of PAC dosages on pH after 2 hours settling; Figure 7 (b) The effects of PAC dosages on electrical conductivity after 2 hour settling

3.6 Microorganism measurements in treated water

As described in Table 3, the results of microorganism measurements showed that no coliforms were detected after coagulation. Although the bacterial colony number (BCN) has increased slightly after coagulation by the soils, it still meets the China National Standard for *Drinking Water Quality* (GB 5749-2006), which requires $BCN < 100$ CFU/mL (There is no standard of BCN in drinking water standards of Ethiopia or WHO). In addition, soil B introduced much more bacteria than soil A.

Table 3 Bacterial colony number (BCN) and coliforms

Water samples	Undiluted		Diluted 10 times	
	BCN (CFU/mL)	Coliforms (CFU/mL)	BCN (CFU/mL)	Coliforms (CFU/mL)
Distilled water	<30	0	<30	0
Kaolin suspension	<30	0	<30	0
Water purified by soil A	31	0	30	0
Water purified by soil B	84	0	40	0

4. Conclusion

In this study, we investigated the characteristics of two local natural soils from Ethiopia for coagulation in drinking water treatment. It was found that pH, TOC and the combination of poly-aluminum chloride (PAC) influence the performance of these soils. Both soils achieved high turbidity removal efficiency (>84.3%) at a wide initial pH range (5~9). When the initial pH varied from 5 to 9, the final pH of water treated by soils ranged from 7.4 to 8.5, which meets the drinking water standards of Ethiopia ($6.5 < pH < 8.5$). Both soil A and soil B could help improve the turbidity removal rate when 0.5 mg. L^{-1} or 1 mg. L^{-1} PAC was used as coagulant, and PAC at these concentrations also play a leading role and have positive influence during the combination use process. The electrical conductivity of combination of soil A and PAC meets the drinking water standards of WHO. The final turbidity of water treated by 5 g. L^{-1} soils could meet the drinking water standards of Ethiopia (5 NTU) after settling for more than 10 hours. Because the soil coagulants are used for point-of-use water treatment, 10 hours settling is acceptable. In addition, the microorganism tests indicated that the total bacteria number of treated water increased from less than 30 CFU to 40 CFU (soil A) or 81 CFU (soil B).

B), which still meet the drinking water standards of China (≤ 100 CFU).

Acknowledgment

This research was supported in part by Fundamental Research Funds for the Central Universities (No. 0400219312). The work was also partially supported by Science and Technology Commission of Shanghai Municipality (No. 15230724300).

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