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Investigation of the process of adsorption of heavy metals in coastal sands containing micro-plastics, with special attention to the effect of aging process and bacterial spread in micro-plastics

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Abstract: he chief purpose of this study is to investigate the process of adsorption of heavy metals in sands containing microplastics due to aging and bacterial culture. For this purpose, first, the experiment's conditions were determined by reviewing previous studies and examining the effects of factors on the duration of bacterial culture and UV radiation. Finally, the test conditions were determined as follows: 25 g of adsorbent in 250 ml solution containing 50 mg/l of lead, cadmium, copper, zinc, chromium, and nickel, 750 micrograms of microplastic, bacterial culture time two days, aging time with UV light 14 days. Results of the study show that the addition of virgin microplastics has little effected on increasing the adsorbent strength, except in the case of nickel which reduces adsorption strength. The aging process increases the absorption of all studied metals by up to 60%. Bacterial culture without an aging process reduces the absorption of nickel and cadmium. Simultaneous use of bacterial culture and aging increases the adsorption power by up to 80% for all metals.

Introduction

Plastic debris found in seas and oceans worldwide is often composed of plastic particles smaller than 5 mm. Ingestion of plastic waste by animal species such as mammals, turtles, fish, or seabirds has caused concern among scientists, managers, policymakers, and the public (Sharma et al., 2017). Plastics themselves contain toxic additives, such as emollients, flame retardants, and antibacterial agents that are released into the ocean and cause pollution (Manafi and Nasab 2017). Also, contaminants such as polycyclic aromatic hydrocarbons and polychlorinated biphenyls can be absorbed onto the plastic surface (Rochman et al., 2013). Then, they enter the animal's body through food consumption and pose many threats to the environment (Zhang et al., 2018 (a)). These threats are due to the effective photosynthesis of seaweed (Sundbaek et al., 2016), hatching, and the reproduction of certain marine organisms (Ding et al., 2019) and lead to the death of certain marine organisms. Contaminants carried by or placed on microplastics can travel globally, poisoning marine organisms, enriching organisms, and further transporting them through the food chain (Peixoto et al., 2019).

In 2015 study of Turner and Holmes PE particles for the adsorption of metal ions Cr, Cu, Ag, Cd, Hg, Ni, Co, Pb, and Zn in Freshwater were used. The results showed that the difference in ion concentration between the liquid phase (metal solution) and the solid phase (PE particles) was the main factor affecting the adsorption and the metal ion concentration reached equilibrium. In aged particles, due to the oxidation and weathering of the particles, the surface morphology of the particles changes, in which it is easy to obtain an electric charge and absorb metal ions to achieve charge balance (Turner and Holmes, 2015).

The process of adsorbing metals to plastics is very diverse and complex and can be said to have remained undiscovered. It seems that weathering processes affect the adsorption capacity more than plastic. Heavy metals, such as copper and zinc, have been reported on waterborne or sediment microplastics, including cadmium, lead, and bromine (Brennecke et al., 2016, Massos and Turner, 2017). In laboratory studies, the secretion of zinc in the intestines of artificial earthworms associated with microplastics was about twenty times higher than that of sediment. Hence, the theory is that microplastics increase the bioavailability of metals in lower feeders. Hodson et al., 2017). Various studies have reported that microplastics can carry Cr at concentrations of 19–7970 ng/g (Holmes et al., 2012, Vedolin et al., 2018).

Various studies have shown that the aging process of microplastics has significant effects on their absorption. A study by Mao et.al in 2020 showed that when PS is exposed to ultraviolet light in different conditions, it has various characteristics (Mao et al., 2020). Also, the results of this study show that the aging sequence of PS functional groups in air and water is known. An isothermal adsorption model showed that aging significantly increases the adsorption of heavy metals by PS. Also, in the aging process, microplastics change color, even crumbling (Müller et al., 2018).

Bacterial communities have significant functions in soil due to their contribution to the nutrient cycle, plant symbiosis, decomposition, and other ecosystem processes. The coexistence of microplastics and bacteria in the soil causes changes in all three of them (Viršek et al., 2017). Microbial inhabitants on the surface of plastics are called „Plastisphere" and researchers have recently reported that microorganisms can form biofilms on microplastics surfaces in aquatic environments and microplastic as a chemical carrier (Mincer et al., 2016). Human pathogens associated with floating microplastics (polyethylene, polypropylene, and polystyrene) have been reported from the North Sea and the Baltic. A published 2016 research stated that the bacteria used microplastics as potential carriers for their dispersal (Kirstein et al., 2016).

Erythrobacter is one of the most important groups of bacteria considered by researchers in the field of microplastics. This bacterial family can use microplastics as a carrier and spread them widely (Curen and Sandric, 2019). *Erythrobacter* is a rod- shaped gram-negative bacterium and usually uses a mobile flagellum. It may branch out (Park et al., 2020). The cells are orange, contain Bchl and carotenoids, and proliferate in the binary division (Fang et al., 2019). No growth occurs anaerobically in light. Ribulose carboxylase diphosphate is not detected. No fermentation and disinfectant activity occur (Jiang et al., 2018).

In this study, we seek to confirm these ideas: The amount of heavy metal adsorption on the sand change in the presence of microplastics. UV exposure changes the adsorption of heavy metals. Also, the presence of certain bacteria in the sand increases the microplastic capacity for heavy metal adsorption. Bacteria and UV rays have a synergistic effect on the process of adsorbing heavy metals.

Methods and materials

The sand samples were collected from the southern shores of the Caspian Sea in Gilan province. Sand samples were taken from the surface to a depth of 7 cm using a sampling tool: Edelman, bott, coarse sand, bay, Ø 7 cm, and each sample weighs 250 grams. An extraction protocol is performed on samples to extract the organic materials and plastic contaminants. With an initial wet weight of 250 g, the resulting dry weight and weight differ from sample to sample (224.2 g to 228.9 g). A temperature of about 60°C and a drying time of 48 hours were required to achieve dry weight. The samples were added to the $ZnCl_2$ (1.8 g/cm^3) solution (Vogler et al., 2019), and stirred for 10 minutes, then left to stand for 2 hours to allow the plastics to float on the solution. Afterward, the plastics were removed from the solution surface, and the sediments were washed with distilled water (Rodrigues et al., 2020).

The ultraviolet lamp used in this research is Mineralight UV Germicidal, Daigger Scientific. Also, *Erythrobacter* longus DSM 6997 and *Erythrobacter* litoralis DSM 8509 were purchased from the DSMZ culture collections. Bacteria were cultured in vitro at 25°C on dip slide agar plates. The concentrations of heavy metals in the solution after absorbing were determined using atomic absorption spectrometry of Unicom 939.

After determining the different conditions for performing the experiments, the various tests were carried out as follows:

1. 25 g of adsorbent (sand) was prepared in a solution of 10, 50, 100, and 200 mg/l of lead, cadmium, copper, zinc, chromium, and nickel. These values represent the known levels of heavy metals in seawater and beaches (Türkmen and Budur 2018). The solution was stirred for one hour, and after that filtrated and analyzed by atomic absorption, the amount of residual heavy metals was determined. Eventually, it was determined the percentage of adsorbed metals.
2. The same conditions as in the previous step were repeated, except that in this step 750 μg of fresh microplastics was added to the adsorbent.
3. The experiments' conditions were the same as before, except that the adsorbent compound was exposed to ultraviolet radiation for 14 days.
4. At this stage, the test conditions were similar to the second stage, except that the adsorbent was placed under bacterial culture for two days.
5. In the last step, the test conditions were similar to the second step, except that the adsorbent was first cultured with bacteria for two days and then exposed to ultraviolet light for 14 days.

Results and discussion

Determininge the amount of adsorbent and heavy metals

To determine the amount of adsorbent and heavy metals, various articles that had researched the adsorption of heavy metals on natural materials were reviewed, and the initial adsorption conditions were determined as follows.

For each experiment, a 250 ml solution containing 50 mg/l of each heavy metal is absorbed on 25 g sand. The pH of the solution is 5.5, and the temperature of the solution is 25°C. The adsorption process uses a stirrer for one hour. The results of articles reviewe the articles are presented in Table 2.

Table 1. XRF results of sand samples

Major Elements (wt%)										Wt%	Wt%
SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	Total	LOI
72.05	0.26	14.71	2.48	0.05	1.02	3.86	2.90	2.62	0.05	100	1.81

Determininge the amount of microplastics

Some researche has been carried out to understand the types and amount of microplastics in the soil. The results of this study are presented in Table 3. As can be seen in the table, the amounts and types of pollutants vary highly from place to place.

To select the type and amount of microplastics, we based the research in the sampling area of this study. Polystyrene, Polyethersulfone, Polypropylene, Polyethylene, and Polyethylene terephthalate granules have been used to make microplastic samples based on the research done by Mehdi

Table 2. Review of articles on the absorption of heavy metals on natural materials

adsorbent	Adsorbent (g/l)	Pb (mg/l)	Cd (mg/l)	Cu (mg/l)	Zn (mg/l)	Cr (mg/l)	Ni (mg/l)	Reference
Clay	25	100	55	30	30	25	–	(Bradl, 2004)
Zeolite	37–150	–	–	20	120	–	–	(Motsi et.al. 2009)
Zeolite synthesized from fly ash	250	100	100	100	–	–	100	(He et.al. 2016)
Bentonite	120	300	300	–	–	–	–	(Andini et.al. 2006)
Modified bentonite	10	–	–	200	200	–	–	(Tohdee and Kaewsichan, 2018)
Bentonite	30	–	–	70	70	–	–	(Kubilay et.al. 2007)
Modified bentonite	33	20	–	20	–	–	–	(Kakaei et.al. 2020)
Goethite and hematite nano-photocatalysts	0.3	–	–	50	–	–	–	(Chen and Lee, 2010)
Scoria	60	200	110	60	60	–	–	(Kwon et.al. 2010)
Scoria	5–500	–	–	–	1000	–	–	(Ponce-Lira et.al. 2017)
Scoria	30–70	–	400	400	–	–	–	(Seyfi et.al. 2015)
Clay	1	–	–	100	100	–	–	(Veli and Bilge, 2007)
Clay	0.5	–	50	–	–	–	–	(Rao and Kashifuddin, 2016)
kaolinite and montmorillonite	2	50	50	–	–	–	50	(Gupta et.al. 2008)
illitic clay	10	50	50	–	–	–	–	(Ozdes et.al. 2011)
Sodium tetraborate--modified Kaolinite	0.2	60	60	–	–	–	–	(Unuabonah, et.al. 2008)
Sand	10	50	–	50	50	50	–	(Awan et.al. 2003)
Chitosan-coated sand	2.5	100	–	100	–	–	–	(Wan et.al. 2010)
Iron oxide-coated sand	1	–	–	30	–	–	30	(Boujelben et.al. 2009)

Table 3. Amount and types of micro-plastics in soil and sand

Background	Type of MP	Amount of MP (mg/kg)	References
Soil	PE	750	(Rillig, 2012)
Soil	–	300–67000	(Rillig, 2018)
Clay Soil Sandy Soil Loess Soil	LDPE- PP	140 160 150	(Zhang et.al. 2018(b))
Agricultural soils	DPE-Polyester-PVC-Nylon-Acrylic	28000	(Corradini et.al. 2019(b))
Soil	LDPE-PET-PVC	60000	(Corradini et.al. 2019(a))
Industrial soils	PVC (>80%), PE, PS	300–67,500	(Fuller and Gautam, 2016)
Floodplain soils	PE-PS-SBR-PVC	55.5	(Scheurer and Bigalke, 2018)
Vegetable field soils	PAEs	18.8	(Li et al., 2016)
Arable soils	**HP**	7.1	(Hu et al., 2003)
Fertilized and cultured field soil	DEHP-DBP	0.014–2.5	(Vikelsøe et al., 2002)
Soil	DEHP-PBDEs	0.025–1.60	(Rhind et al., 2013)
Soil and street dust samples	**HP**	0.19–2.12	(Škrbića et al., 2016)
Soil	4-nonylphenols-phthalates-PCB	0.042–0.099	(Gibson et al., 2005)

Nia et al. (2019). According to the study, more than 70% of microplastics on the shores are in the form of fragments and films. Hence, the granules used were converted into films and fragments. A Mill Powder Tech Solutions RT series was used to convert into a fragment. The method presented in 2016 research is used to prepare microplastic film (Cole 2016). Polystyrene, Polyethersulfone, Polypropylene, Polyethylene, and Polyethylene terephthalate fibers (10–30 µm diameter) were aligned. Then, they were embedded in the water-soluble freezing agent and sectioned (30–100 µm length) using a cryogenic microtome. A composition was prepared from various prepared microplastics with an amount of 3×10^5 micrograms per kilogram of sand.

Determining the optimal time for bacterial culture

The culture time of bacteria was 1 to 4 days, and the time of exposure to ultraviolet rays was 1 to 14 days. Thus, a 250 ml solution containing 25g of sand, and 750µg of microplastic was subjected to various times for bacterial culture and ultraviolet conditions. Then the process of adsorbing 12.5 mg of each metal contaminants was performed for one hour of stirring. The results of these experiments are shown in Figure 1.

Lead adsorption in bacterial culture with a gentle slope has increased from the first to the fourth day. But in the aging process, from the 13th day onwards, the upward trend occurreds with a very steep slope. The amount of cadmium adsorption in the aging process with a gentle slope increased from the first to the fourteenth day. But in the process of bacterial culture on day 2, a peak was created in the chart. The copper adsorption amount in the aging process with a gentle slope increased from the first to the fourteenth day. Nevertheless there is no evident change in the bacterial culture process. The amount of chromium adsorption in the aging process with a gentle slope increased from the first to the fourteenth day. However, in samples containing two-day bacterial cultures, they show an increase in absorption from the seventh day of aging. The amount of zinc adsorption in the aging process with a very slight slope increased from the first to the ninth day, but in samples containing two-day bacterial culture, from the ninth day of aging it shows a sudden increase in absorption. The nickel uptake process shows a similar procedure to cadmium, except that the sample contains bacteria with three- and four-day cultures, up to the sixth day of the aging process, followed by the downward trend, and then the downward trend to the ninth day and again the upward trend occurred.

In the adsorption of cadmium, chromium, zinc, and nickel metals, a mutation in the adsorption occurs during culture for two days. Therefore, for the main tests, the bacterial culture

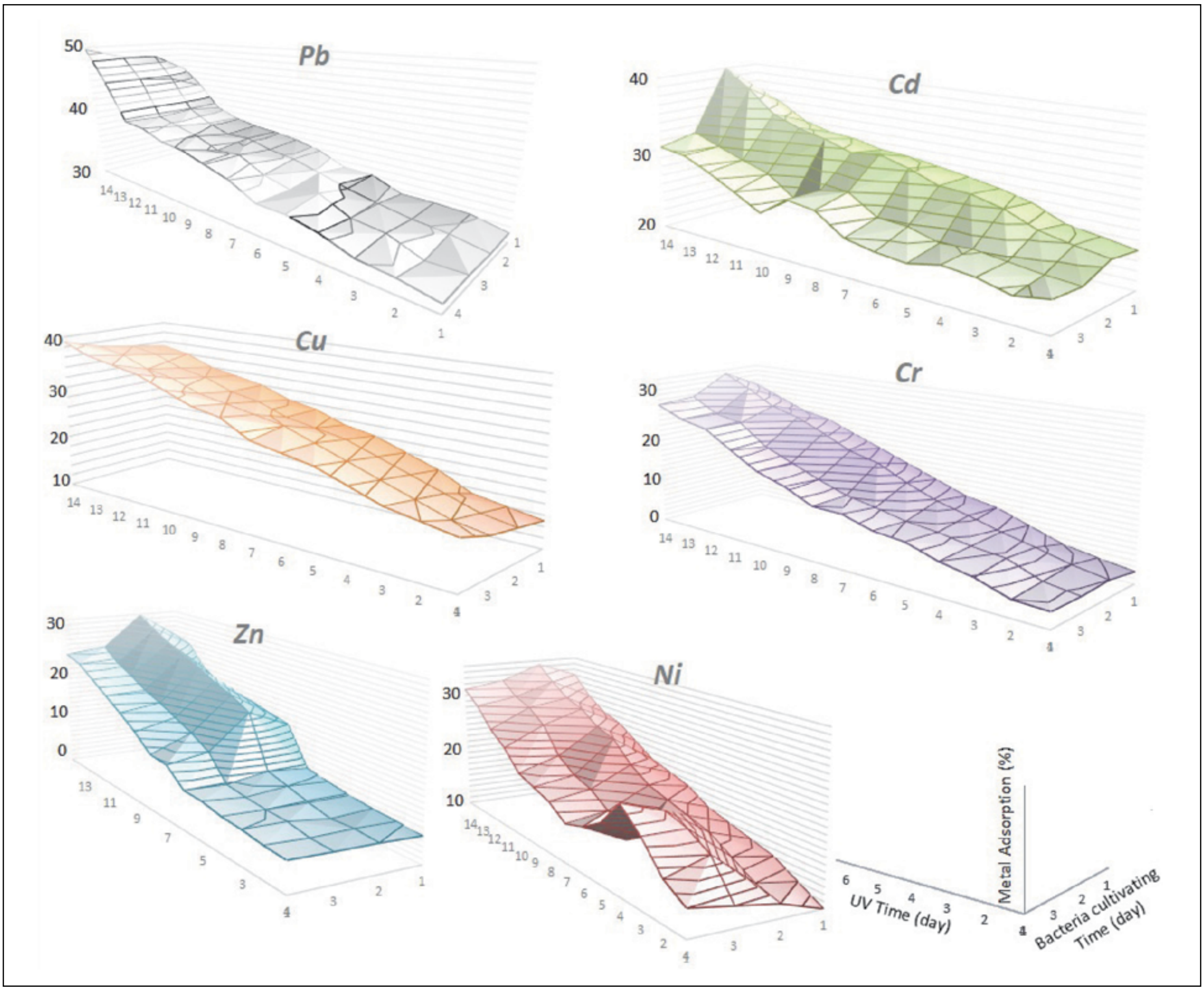

Fig. 1. Results of heavy metal adsorption during bacterial culture for 1 to 4 days and UV exposure time from 1 to 14 days

time of 2 days was selected as the test conditions. Continuous exposure to ultraviolet radiation has increased the adsorption of heavy metals. According to this explanation and figure 2, 14-day UV exposure test conditions was determined for the rest of this study.

After determining different conditions for performing the experiments, the various tests were done as mentioned in the method and materials. Figure 3(A) shows the results of stage 1 of this study. The highest adsorption on sand belongs to lead and the lowest to chromium. The results of step 2 can be seen in Figure 3(B). In this case, the adsorption values have changed, but the general trend of the graphs does not show much change. In conditions of stage 3, the amount of metal adsorption has increased dramatically, and previous trends show noticeable changes (Figure 3(C)). Compared to the third stage, there is a noticeable decrease in the metal adsorption amount in stage 4 (Figure 3(D)). The step 5 conditions, while disrupting the overall trend of the graphs, have caused a significant increase in metal adsorption Figure 3(E)).

Figure 4 shows that for all metals and at every concentration, the increase in fresh microplastics has little effect on metal adsorption. It is only in nickel that the addition of microplastics has reduced the absorption of the metal. Exposure to ultraviolet light increases the absorption of heavy metals at all concentrations. Interestingly, culturing the bacteria without exposure to ultraviolet light reduces the adsorption capacity of the sand and microplastics. But the combination of ultraviolet rays with bacteria increases the absorption power of sand and microplastics significantly.

Figure 5 shows the change in the percentage of adsorption in each state from 2 to 5 compared to state 1. The most observed change is related to the fifth state, which means adsorption on sand and microplastics under the influence of bacteria and ultraviolet rays. Until a few years ago, plastic particles were considered relatively inert compared to heavy metals (Ashton et al., 2010), however, recent studies show that these particles can metal at a rate similar to that in sediments. And suspended particles are observed to accumulate (Duarte et al., 2014).

Adding fresh microplastics to the sand reduces the amount of adsorption on nickel. Also, this conclusion is stated in a research in 2019, where the adsorption of 134Cs and 85Sr in size range of 50 to 100μm microplastic sediments was generally 2–3 times lower than the adsorption on sediments (Johansen et al. 2019).

The exposure to ultraviolet radiation has increased the absorption of all metals by about 20 to 60 percent. Differences in metal adsorption between virgin and coastal polyethylene pellets were observed in similar laboratory pellet suspension experiments under marine and river conditions (Holmes et al., 2014).

Also, bacterial culture without ultraviolet radiations has reduced the absorption of nickel and cadmium compared to the adsorption on the sand. A similar result is found in another study showing that the concentration of metals on plastics

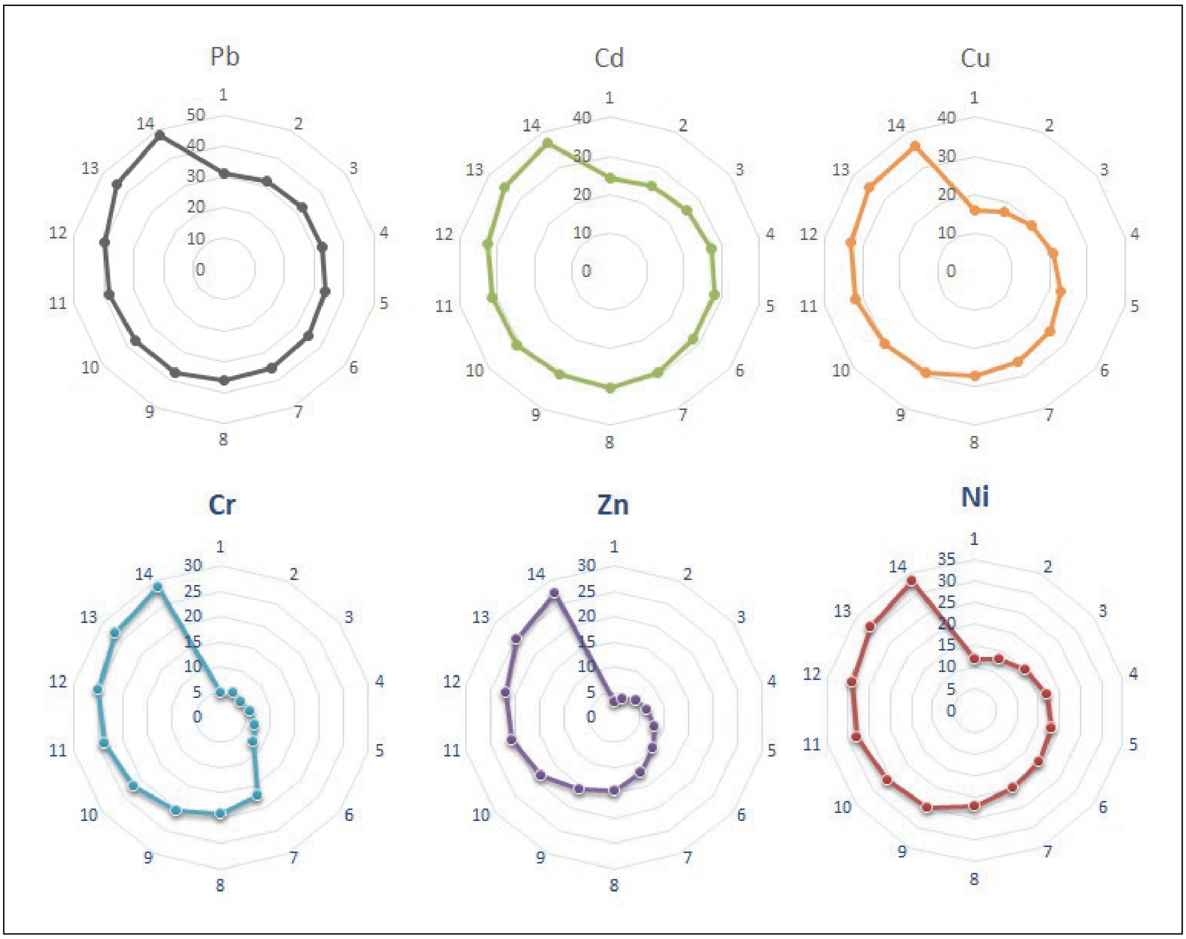

Fig. 2. The trend of increasing the adsorption of heavy metals by increasing the exposure of the adsorbent to UV exposure

was 800 times higher than the surrounding seawater, which is indicated by the partition coefficient and there is a significant difference in the concentration of metal compared to PVC parts and PS grains before testing. Because these old pellets are in higher degradation, the adsorption capacity may increase, as is suggested by Endo et al. (Endo et al. 2005).

Simultaneous exposure of adsorbent to bacteria and ultraviolet light has resulted in an increase of about 60–80% for all metals. In the case of nickel, the effect of bacteria is not very visible. It means that the UV exposure alone or with bacteria does not make much more difference in the heavy metals absorption amount. In the case of metal, the effect on bacteria and UV was almost the same, but the simultaneous presence of these two factors did not cause a synergy in the adsorption amount. Regarding cadmium metal, although the effect of bacteria alone has reduced the amount of adsorption, the presence of bacteria and UV have had a synergistic effect on each other. The results of a 2020 study show a high interaction between the degree of plastic aging and the adsorption capacity to heavy metals (copper and zinc). Meanwhile, external conditions, including temperature and pH, showed a high effect on the adsorption behavior (Wang et al. 2020).

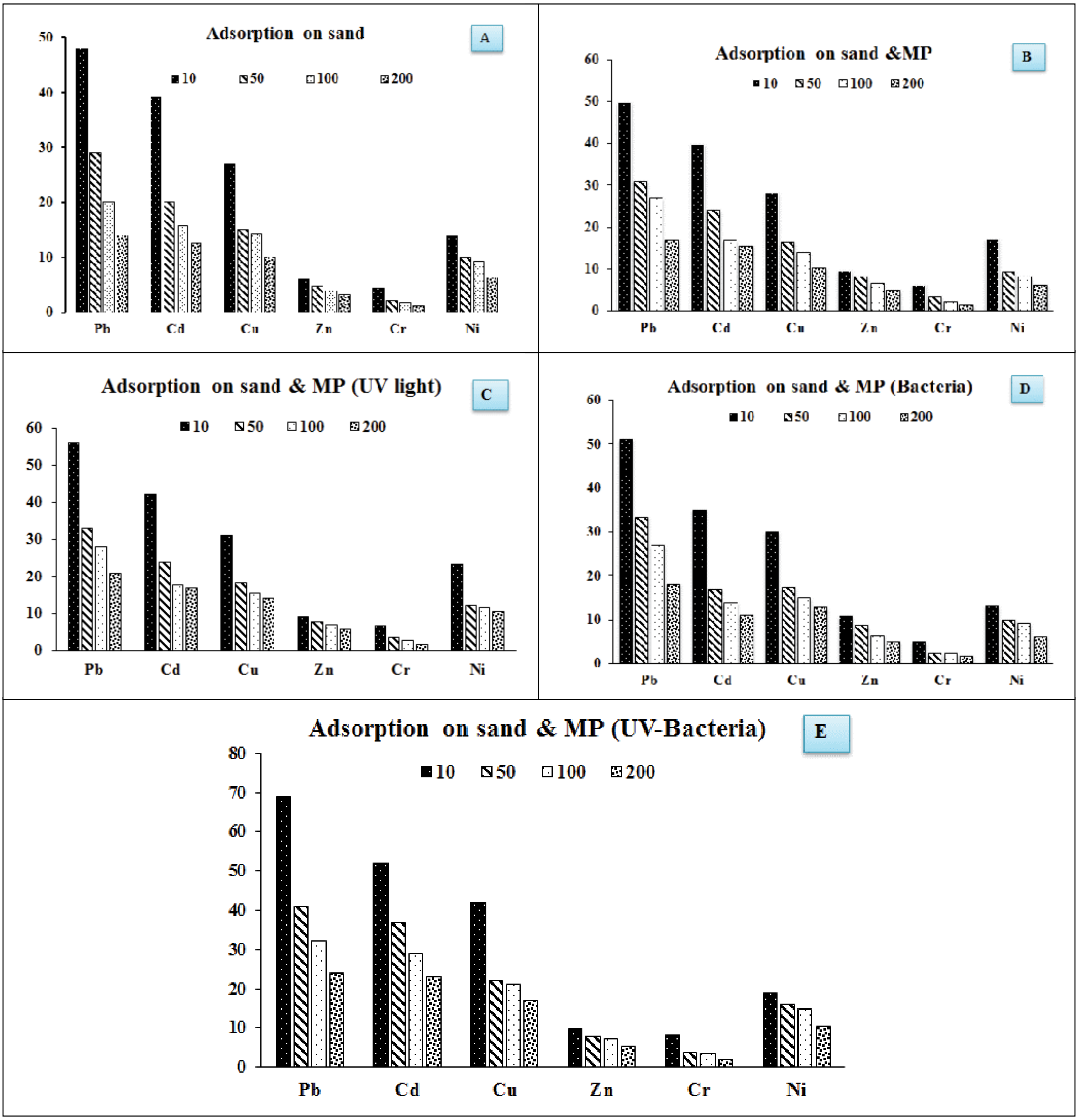

Fig. 3. Adsorption of heavy metals on A) Sand, B) Sand and micro-plastics C) Sand and micro-plastics exposed to UV D) Sand and micro-plastics containing bacteria E) Sand and micro-plastics containing bacteria exposed to UV

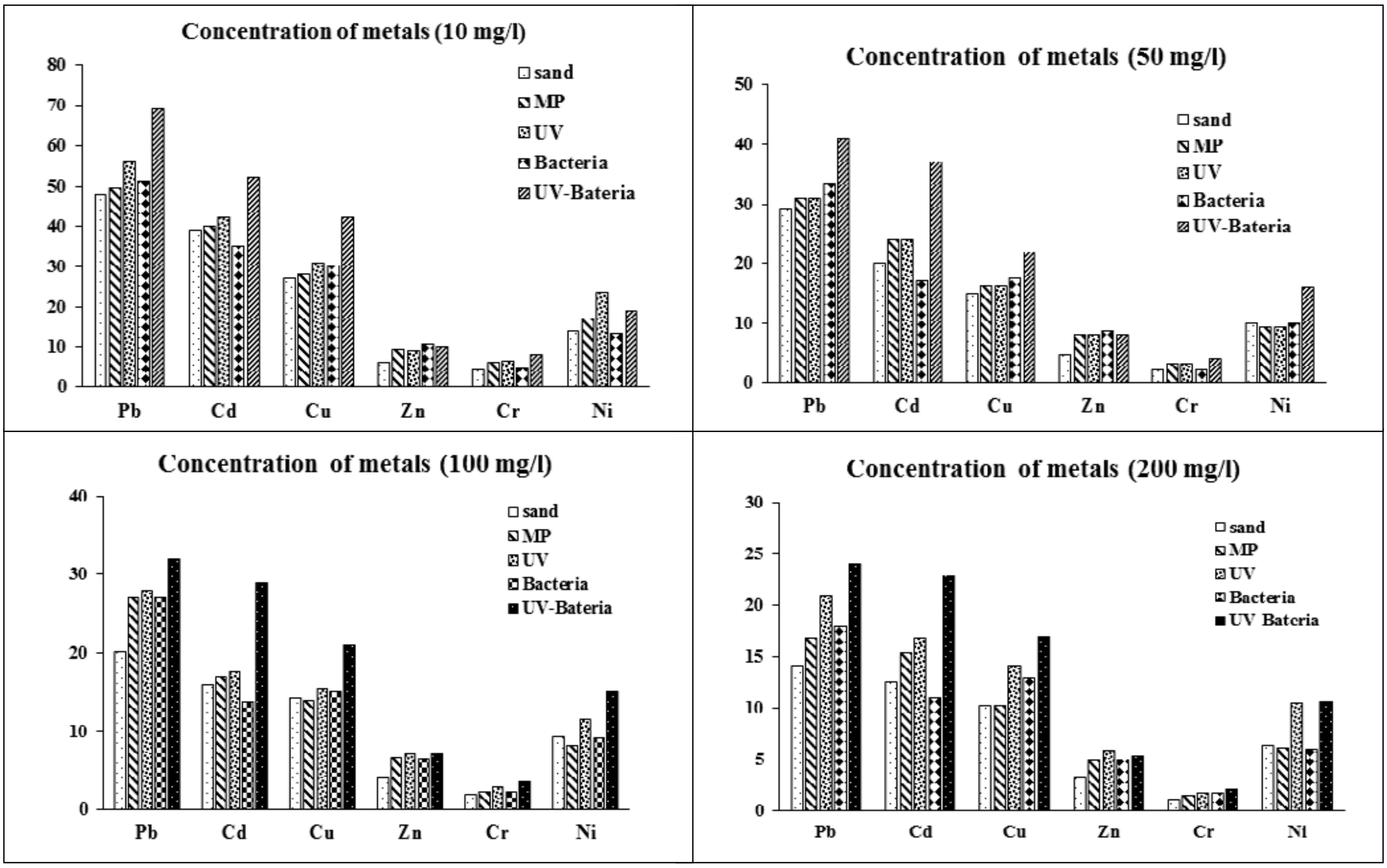

Fig. 4. Percentage of adsorption of heavy metals on various compounds of sand and micro-plastics, under UV irradiation and bacterial culture at concentrations of 10, 50, 100 and 200 mg/l

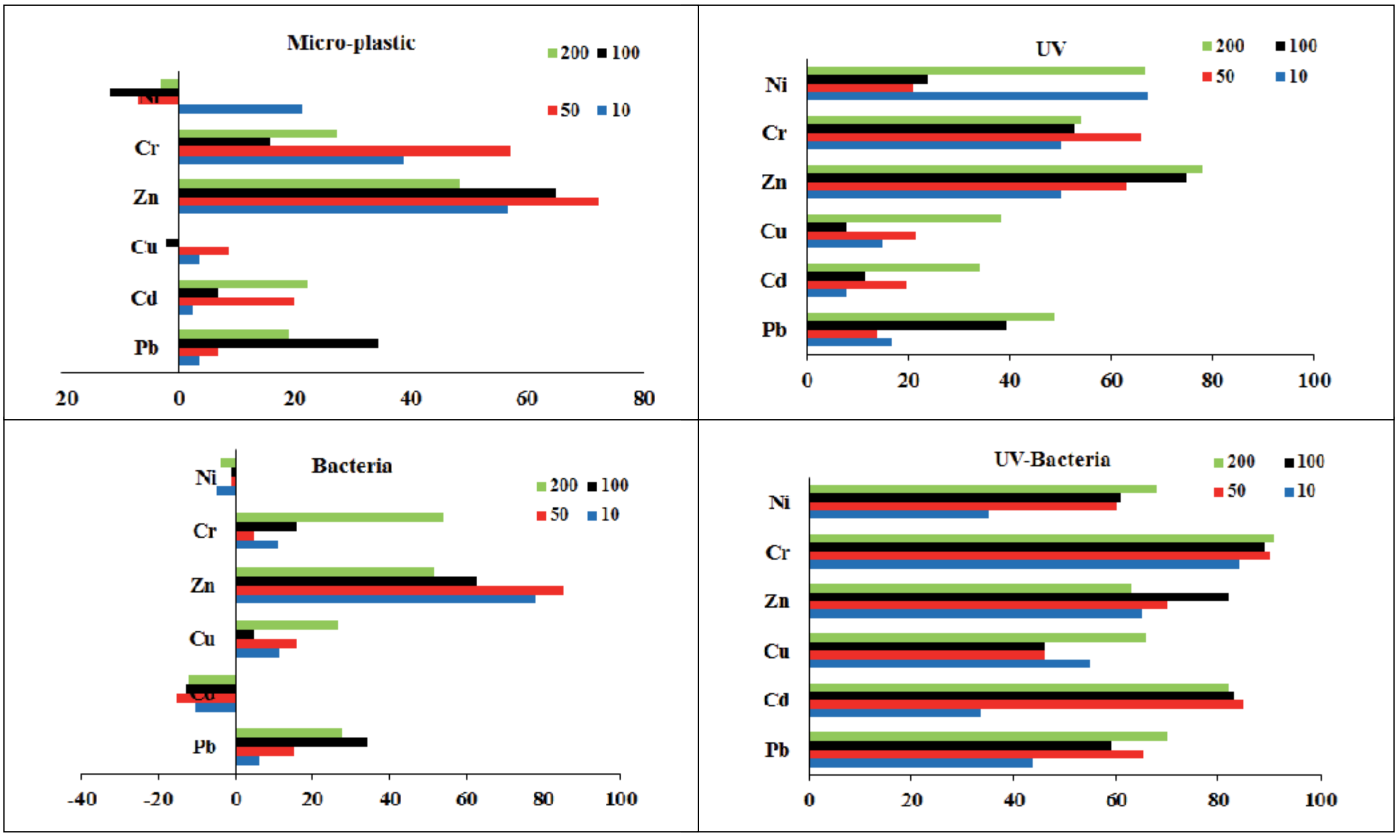

Fig. 5. Changes in the adsorption percentage in different adsorbent states compared to the adsorption mode of heavy metals on sand

Conclusions

In this study, firstly, the primary factors were determined for performing experiments. Absorbent weight, metal concentration, type, and amount of microplastics were determined by reviewing previous studies. Thus, each test was performed with 25 g of sand, 750 µg of microplastics, and 50 mg/l of each heavy metal. The temperature was 25°C, and pH was 5.5. For determining how long bacteria would be cultured in the adsorbent, 1 to 4 days, bacterial culture experiments were performed. Simultaneously, the duration of the aging process from 1 to 14 days was examined. The results showed that the uptake of metals increased and decreased until the second day of bacterial culture. Also, the aging time with UV rays is ever-increasing, and therefore 14 days of the aging process was selected. Under these conditions, various experiments were performed, in which the concentration of metal contaminants was 10, 50, 100, and 200 mg/l, and in five stages pure sand, containing fresh, aged, exposed to bacteria and simultaneously exposed to bacteria and aged microplastics were used as adsorbents.

The results showed that the addition of fresh microplastics had little effect on increasing the adsorbent strength, except the nickel, which reduces adsorption strength. The aging process increases the adsorbent power to absorb all metals by 20 to 60%. Bacterial culture alone decreases the uptake of nickel and cadmium and has the most effect on zinc uptake. Simultaneous use of bacterial culture and aging increased the adsorption power by about 60 to 80% for all metals.

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