Shungite application for treatment of drinking water – is it the right choice?

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ABSTRACT

Shungite is a natural carbon containing material that is widely used in water treatment. Scientific research shows that shungite has good adsorption properties towards various organic compounds and heavy metals, as well as exhibiting antibacterial properties. Unfortunately, at the same time shungite releases various chemical elements into the water, including heavy metals. In this study changes in concentration of various heavy metals during drinking water treatment with one commercial and one non-commercial shungite sample were determined. Also sorption of Cu(II) with initial concentration of 2,500 μg/L onto shungite was investigated. The results showed that various heavy metals like nickel, copper, lead, cadmium, zinc, chromium and arsenic are leaching from shungite into water. Lead and cadmium exceeded the maximum acceptable concentration in drinking water for a few days, but nickel exceeded for up to 2 weeks. At the same time shungite showed good adsorption properties towards copper. Nevertheless, before using shungite in drinking water treatment, it would be advisable to assess the necessity and/or wash shungite with larger volumes of water for a longer period of time than is written in the instructions.

Key words | adsorption, heavy metals, release, shungite, water treatment

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HIGHLIGHTS

- Drinking water was treated with two shungite samples based on the application instructions of the used commercial shungite sample.
- Both shungite samples released various heavy metals like nickel, copper, lead, cadmium, zinc, chromium and arsenic.
- The released nickel exceeded the maximum acceptable concentration in drinking water up to 2 weeks but lead and cadmium for a few days.

INTRODUCTION

Shungite is a natural mineraloid that contains non-crystalline carbon. There are five types of shungite, classified by carbon content. Type I shungite contains more than 98 mass% of glass-like carbon, type II contains 35–80 mass%, type III 20–35 mass%, type IV 10–20 mass%, but type V contains <10 mass% of carbon. Type III shungite is the most widespread and the largest shungite deposits are located in Karelia region, Russia (Melezhika *et al.* 2004; Mosin & Ignatov 2013; Sineva 2014). In addition to carbon, shungite usually contains quartz, aluminosilicates, feldspars and carbonates. Also, various micro impurities can be found in shungite: Fe, Ni, Cu Zn and V, mainly as sulphides,

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sulphates and oxides (Charykova *et al.* 2006; Rafienko & Belimenko 2019). Charykova *et al.* (2006) showed that in addition to these micro impurities type III shungite also contained Cr, Co, Pb and Mn.

There are a large number of patents on shungite application for drinking water treatment (more than 100 results revealed in the patent search engine https://worldwide.espacenet.com with search keywords 'shungite' and 'drinking water') and a wide range of commercial shungite products for water treatment at home are also available (Karelian Heritage 2020). Information on most of these products claims that shungite removes bad taste and odour, organic compounds, heavy metals and bacteria and enriches water with microelements. Indeed, studies show that shungite has good adsorption properties towards various organic compounds (Kalsina & Berjoza 2006; Sineva *et al.* 2007; Skorobogatov *et al.* 2013) and also antibacterial properties (Charykova *et al.* 2006). Fischer *et al.* (2018) concluded that low-carbon shungite (total carbon content 5.4%) could be used as an alternative adsorbent for Zn(II) removal from water. Efremova (2006) showed that porous sorbent prepared from shungite rock can adsorb Cd(II), Pb(II), Zn(II) and Mn(II) in dynamic conditions. Nevertheless, Charykova *et al.* (2006) showed that a large number of chemical elements are leaching from shungite into water, including several heavy metals like Cd, Cr, Cu, Ni, Pb and Zn. After 3 days of shungite contact with tap water, many elements exceeded the maximum acceptable concentration in drinking water. The authors suggested that the increased concentrations of some heavy metals could be the reason for the antibacterial properties of 'shungite water' (Charykova *et al.* 2006). Some of the elements in large quantities are toxic for humans, therefore in the few websites about shungite it is written that it is advisable to drink just one or two glasses of 'shungite water' due to the presence of heavy metals.

The aim of this study was to determine the changes in concentration of various heavy metals (Ni, Pb, Zn, Cd, Cu, Cr, As, Al) during the application process of drinking water treatment with one commercial and one non-commercial shungite sample. As Cu(II) can still be found in drinking water due to corrosion of copper pipes, the sorption of artificially increased concentrations of copper ions in drinking water was also investigated.

MATERIALS AND METHODS

Materials

Two shungite samples with particle sizes in the range of 1–3 mm were used. Sample Com is a commercial product 'ШУНГИТ PREMIUM' for water purification at home (ШУНГИТ PREMIUM КЛАССА 2020) and sample SH is from the fields of Karelia region (Russia). For all experiments Evian® natural spring water was used as model of drinking water (in further text – water). Copper sulphate monohydrate (≥98%) was purchased from Sigma-Aldrich (St. Louis, MO).

Characterization of shungite samples

Specific surface area (SSA) was determined by nitrogen gas adsorption performed with QuadraSorb SI (Quantachrome Instruments, Boynton Beach, Florida). SSA was calculated according to the BET method. Before the analysis all samples were degassed at 300 °C for 3 h.

Carbon content was determined by an element analyzer Vario Macro CHNS (Elementar Analysensysteme GmbH, Germany).

Preparation of 'shungite water'

The procedure was based on the application instruction inside of the commercial shungite package. The first step written in the instruction is to wash shungite several times with water. Therefore, 10 g of shungite was poured in a beaker, agitated with 200 mL of water for 2 min and then decanted. This procedure was repeated five times. The last decanted water was free from shungite dust particles. The first decanted water was filtered and used for chemical analysis (sample SH-1x and Com-1x). The procedure for sample preparation is shown in Figure 1. The washed shungite samples were mixed with 100 mL of water (shungite: water mass ratio was 1:10) and left still. According to the instructions, after 2–3 days the water is ready for use and every time a certain amount of water is removed for application, the same amount of untreated water is added to shungite. Therefore, on the third day 50 mL of water was removed from the container and the same amount (50 mL) of fresh water was poured back on the shungite. This

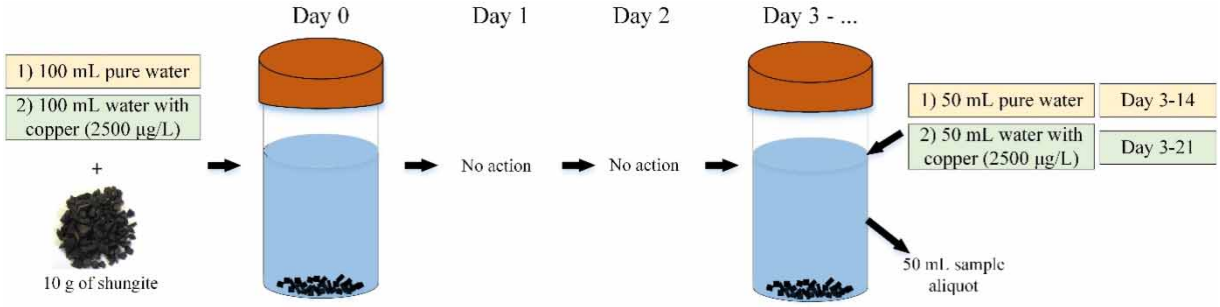

Figure 1 | Procedure of preparation of 'shungite water' samples (1) and samples for adsorption experiments of copper (2).

procedure was repeated every day for the next 14 days. The container with shungite and water was stirred for a few seconds once every day. The chemical analysis was performed for samples removed on the 3rd, 5th, 7th, 11th and 14th day of the experiment.

Adsorption of copper

The initial concentration of Cu(II) ions in water was 2,500 µg/L, which is slightly higher than the maximum acceptable concentration in drinking water (2,000 µg/L) according to the Council Directive 98/83EC. The procedure for the adsorption experiment was equivalent to the preparation of 'shungite water' described above and shown in Figure 1. Chemical analysis was performed for samples removed on the 3rd, 5th, 7th, 11th, 14th, 17th and 21st day of the experiment.

Analysis of water and 'shungite water'

The chemical analysis of samples was conducted at the Latvian Environment, Geology and Meteorology Centre. The concentration of Ni, Pb, Cd, As and Cr was determined according to ISO 15586:2003 standard using an electrothermal atomic absorption spectrometer Varian SpectrAA 880Z (Varian, Palo Alto, California). The concentration of Zn, Cu and Fe was determined according to ISO 8288:1986, Ca and Mg according to ISO 7980:2000, Al according to ISO 12020:2005 and K according to ISO 9964-3:1993 standards using a flame atomic absorption spectrometer Varian SpectrAA 880 (Varian, Palo Alto, California). Only those results equal to or higher than the quantification limit (QL) are shown with the expanded uncertainty (±) with 95% confidence level. The results which were below the method detection limit (MDL) are marked with '<'. For each element, QL and MDL can be different.

Total organic carbon (TOC) and dissolved organic carbon (DOC) analysis were performed for water samples after the 3rd and 5th day of contact with shungite. The samples were prepared due to LVS EN 1484:2000 standard and analysed with FORMACS^{HT} TOC/TN Analyzer (Skalar, Breda, The Netherlands).

RESULTS AND DISCUSSION

Release of heavy metals

Shungite SH has slightly higher carbon content but six times lower SSA than shungite Com (Table 1) which can lead to lower sorption properties. According to the shungite classification in Melezhika *et al.* (2004), SH belongs to type-II but Com to type-III shungite.

The results in Table 2 show that after the first washing (2 min of contact with shungite) the concentration of heavy metals like Ni, Cu, Zn and Cd is significantly increased. The highest increase can be observed for nickel from both samples. From these results we can conclude that these heavy metals are released in high concentrations in water and the washing procedure is mandatory not just to remove the small particles (dust), but also to get rid of heavy metals to avoid contamination of the drinking water intended for consumption.

Table 1 | Carbon content and SSA of shungite samples

Sample	Carbon content, %	SSA, m^2/g
SH	39.3 ± 0.4	1.3 ± 0.1
Com	31.6 ± 0.3	7.9 ± 0.2

Table 3 shows that after 3 days in contact with both washed shungite samples, the water contained increased levels of Ni, Cu, Pb, Cd, Zn and As, compared to pure water (Table 2). However, only nickel, cadmium and lead (for sample SH) exceeded the maximum acceptable concentration (MAC) in drinking water (Table 4). On the 5th day of exposure, the concentrations of released heavy metals had decreased rapidly and were below MAC, except for nickel. Similar results were obtained for water with increased copper concentration after exposure to shungite (Table 5). Here, after 3 days the water also contained increased levels of several heavy metals (Ni, Cu, Pb, Cd, Zn and Cr), where only cadmium and nickel exceeded MAC, but on the 5th day of exposure only nickel exceeded MAC. Compared with pure water, the increase of Ca, Mg, Na, K and As is negligible and does not exceed MAC.

Table 2 | Chemical elements in water before and after the first washing of shungite

Element (MDL; QL)	Water	SH-1x	Com-1x
Ni (1.2; 4), μg/L	<0.9	660 ± 60	175 ± 16
Cu (0.3; 0.9), μg/L	0.3	36 ± 5	35 ± 5
Pb (0.4; 2), μg/L	0.4	1.1	1.0
Zn (10; 30), μg/L	13	153 ± 29	36 ± 7
Cd (0.02; 0.05), μg/L	<0.007	3.5 ± 0.6	1.0 ± 0.2
Cr (0.2; 0.5), μg/L	<0.2	0.3	0.3
As (0.2; 0.6), μg/L	<0.2	0.4	0.3
Al (1; 3), mg/L	<1	<1	<1
Ca (0.2; 0.6), mg/L	69 ± 10	76 ± 11	72 ± 10
Mg (0.1; 0.4), mg/L	26 ± 2	27 ± 2	26 ± 2
Fe (0.04; 0.15), mg/L	<0.04	<0.04	<0.04
Na (0.2; 0.5), mg/L	5.4 ± 0.3	5.9 ± 0.4	5.7 ± 0.3
K (0.1; 0.4), mg/L	1.0 ± 0.1	1.1 ± 0.1	1.3 ± 0.1

Table 3 | Concentration of various metals in water after shungite treatment (elements exceeding MAC are underlined)

	SH		Com	
Element	3rd day	5th day	3rd day	5th day
Ni, μg/L	880 ± 80	58 ± 6	1700 ± 150	60 ± 6
Cu, μg/L	4.3 ± 0.6	<0.3	33 ± 4	0.8
Pb, μg/L	211 ± 17	<0.5	5.0 ± 0.4	1.1
Zn, μg/L	129 ± 25	<10	900 ± 170	<10
Cd, μg/L	5.0 ± 0.9	0.022	11 ± 2	0.16 ± 0.04
Cr, μg/L	0.25	<0.2	<0.2	<0.2
As, μg/L	0.6 ± 0.1	0.4	0.3	<0.2
Al, mg/L	<1	ND	<1	ND
Ca, mg/L	76 ± 11	ND	81 ± 11	ND
Mg, mg/L	31 ± 2	ND	38 ± 3	ND
Fe, mg/L	<0.04	ND	<0.04	ND
Na, mg/L	6.1 ± 0.4	ND	6.1 ± 0.4	ND
K, mg/L	2.5 ± 0.3	ND	2.0 ± 0.2	ND

ND, not determined.

Table 4 | Maximum permissible concentrations of heavy metals (MAC)

Element	Maximum acceptable concentration in drinking water (Council Directive 98/83EC), μg/L	Tolerable intake level stated by European Food Safety Authority (EFSA)
Nickel	20	2.8 μg/kg of body weight per day (EFSA CONTAM 2015)
Copper	2,000	for adults 1.3 (women) and 1.6 (men) mg/day (EFSA NDA 2015)
Lead	10	NA (EFSA 2012a)
Zinc	NI (5000[a])	for adults 7.5–16.3 mg/day (depends on gender and phytate intake) (EFSA NDA 2014a)
Cadmium	5	2.5 μg/kg body weight per week (EFSA 2012b)
Chromium	50	NA (EFSA NDA 2014b)
Arsenic	10	NA (EFSA CONTAM 2009)

NA, not applicable; NI, not indicated.
[a]EPA (2018).

Table 5 | Concentration of heavy metals (copper not shown) in water with increased copper concentration after exposure to shungite (elements exceeding MAC are underlined)

	SH		Com	
Element	3rd day	5th day	3rd day	5th day
Ni, μg/L	283 ± 28	212 ± 30	400 ± 40	225 ± 32
Pb, μg/L	2.1 ± 0.2	2.8 ± 0.5	3.3 ± 0.3	0.8
Zn, μg/L	290 ± 70	26	910 ± 210	43 ± 8
Cd, μg/L	7.6 ± 1.7	1.5 ± 0.3	10 ± 2	0.9 ± 0.2
Cr, μg/L	0.5 ± 0.1	<0.2	0.5 ± 0.1	<0.2
As, μg/L	<0.2	<0.2	<0.2	<0.2

Based on the experimental procedure (Figure 1), every day (starting from the 3rd day) 50 mL of the water exposed to shungite was replaced with 50 mL of fresh water, therefore the concentration of heavy metals was diluted twice each day. For example, in Table 3, on the 3rd day the nickel concentration was 880 μg/L and if we assume that during the next 2 days shungite was not releasing nickel, the concentration on the fifth day should be 220 μg/L, but the analysis showed four times lower concentration (58 μg/L). The same observation was found for copper, lead, zinc and cadmium. We believe that most likely this could be explained by precipitation of salts due to various anions released from shungite in the water, such as sulphates, sulphides and carbonates (Turkayeva *et al.* 2017; Rafienko & Belimenko 2019). Another possible reason could be shungite adsorbing back some part of the released metals due to the fact that shungite contains and releases organic matter that was measured as TOC and DOC (Table 6). Organic matter and DOC forms complexes with metal ions, thereby affecting the adsorption/desorption process (Khokhotva & Waara 2010). The pH of Evian water was 7.5 and after exposure to shungite the pH of water samples was in the range of 7.1–7.6, therefore the changes in nickel, copper, lead, zinc and cadmium concentration cannot be connected to precipitation due to pH. In Table 5 this rapid concentration decrease is observed only for zinc and cadmium.

Table 6 | TOC and DOC results

		SH		COM	
Parameter, mg/L	**Water**	**3rd day**	**5th day**	**3rd day**	**5th day**
TOC	0	0	1.09 ± 0.09	0.89 ± 0.08	1.16 ± 0.09
DOC	0	0	0.69 ± 0.06	0.44 ± 0.04	0.65 ± 0.06

Furthermore, the concentration of nickel in water was determined for several weeks throughout all experiments (Figure 2). Figure 2(a) shows the nickel concentration in pure water after exposure to shungite and Figure 2(b) shows the nickel concentration in the experiment with water containing artificially added copper. If we compare both these experiments, the released amount of nickel in water is different for each shungite sample, especially after the first 3 days of exposure. On the one hand this could be affected by the inhomogeneous distribution of soluble nickel compounds, due to the natural origin of shungite. On the other hand, it could be connected to the presence of additional copper (Figure 2(b)) that caused a more gradual nickel release from shungite. According to the application instructions of Com, shungite should be changed after six months of application, meaning that every six months for a week or two the consumer of 'shungite water' will be exposed to increased levels of nickel.

In Figure 2(a) and 2(b) the concentration of released nickel from shungite Com is significantly higher than from shungite SH after 3 days of exposure. On the 5th day the difference is negligible, but in the following days the released nickel from shungite Com is lower than from shungite SH. This could be explained by the fact that shungite Com has higher SSA than shungite SH, therefore Com releases nickel much faster.

The recommended tolerable daily intake (TDI) of nickel has changed over the years. In 2005, the European Food

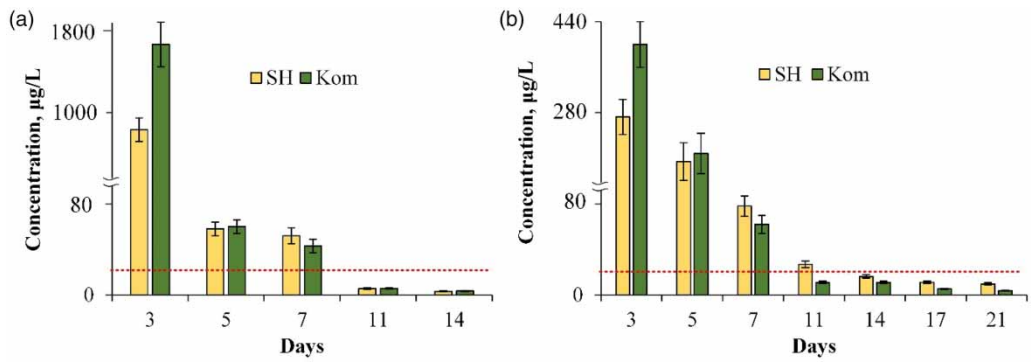

Figure 2 | Nickel concentration in water samples after exposure to shungite: (a) during 2 weeks and using pure water; (b) during 3 weeks and using water with 2,500 μg/L of copper. The red dashed line indicates the MAC of nickel in drinking water.

Safety Authority (EFSA) Scientific Panel on Dietetic Products, Nutrition and Allergies released a scientific opinion related to the tolerable upper level of nickel, where according to scientific investigations and the lack of evidence that Ni is essential for humans it was concluded that it is not possible to establish the TDI for nickel (EFSA 2005). Two years later, in 2007, the World Health Organization (WHO) established the TDI at 11 μg/kg of body weight (WHO 2007). In 2015 the EFSA Panel on Contaminants in the Food Chain released a scientific opinion on the risks of public health related to the presence of nickel in food and drinking water where the TDI of 2.8 μg/kg of body weight was established (EFSA CONTAM 2015). Based on this, for an adult with an average weight of 70 kg the TDI would be 196 μg and to intake such an amount of nickel one would need to drink more than 3 litres of 'shungite water' with an Ni concentration of approximately 60 μg/L (after 5 days according to data presented in Table 3), and even more when the Ni concentration is lower. On the other hand, the major source of Ni uptake is food – cocoa beans and chocolate (Kruszewski *et al.* 2018), beans, seeds, nuts, grains, vegetables, fruits and also products containing them (EFSA CONTAM 2015). The amount of nickel in foods may vary considerably from place to place, due to the different nickel content in the soil.

The biological function of nickel in the human body is still unclear. The highest concentrations of nickel in the human body are found in the nucleic acids, particularly RNA, and it is thought to be somehow involved in protein structure or function. Nickel may play a role, as a cofactor, in the activation of certain enzymes related to the breakdown or utilization of glucose (Kumar & Trivedi 2016).

The most reported effects after acute exposure to Ni are gastrointestinal (vomiting, cramps, and diarrhea) and neurological symptoms (giddiness, headache, and weariness). Ingestion of Ni is able to elicit eczematous flare-up reactions in the skin in Ni-sensitized individuals, but scientific research indicates that it is unlikely that dietary exposure to Ni would result in cancer in humans. Although not all consumed Ni is absorbed from the gastrointestinal tract (1–40% from the amount ingested) (EFSA CONTAM 2015), extra Ni uptake by using 'shungite water' in both the short and long term can cause health problems.

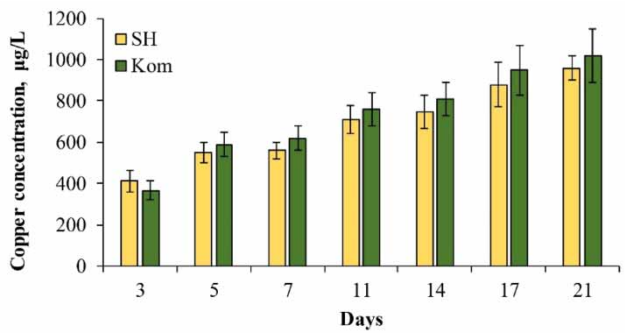

Figure 3 | Remaining copper concentration (μg/L) in water samples during 3 weeks of copper adsorption experiment.

Adsorption of copper

Despite the fact that shungite releases various heavy metals, at the same time it adsorbs copper (Figure 3) from water. The results in Figure 3 show that the remaining copper concentration slowly increases with time, where after 3 weeks the concentration for both shungite samples was approximately 2.5 times higher than after 3 days of exposure. This means that shungite sorption properties towards copper decreases, removing approximately 81–87% of the initial copper concentration after the first 3 days and 40–50% on the 21st day of exposure to both shungite samples. The decrease of copper concentration can also be attributed to the precipitation of copper sulphide due to the possible sulphide ion release from shungite (Rafienko & Belimenko 2019).

As shown in Table 1, shungite SH contains higher amounts of carbon but has lower SSA compared to shungite Com. The obtained results show the carbon content has a direct influence on sorption properties, because overall shungite SH showed slightly higher sorption than Com towards copper.

CONCLUSIONS

In the current study, drinking water treatment with shungite was investigated. The results show that shungite samples release various heavy metals into the water – nickel, copper, lead, cadmium, zinc, chromium and arsenic. Lead and cadmium is released for a short time and exceeded MAC only after the first 3 days of exposure, but nickel is

released for a much longer time and can exceed MAC up to 2 weeks. Increased specific surface area probably accelerates the rate of nickel release from shungite but carbon content in shungite promotes sorption properties.

Based on the obtained data, it would be advisable to give careful consideration to the use of shungite for drinking water treatment. To avoid heavy metal contamination from shungite, prior to application and additionally to the washing procedure written in the instructions, shungite should be washed with a large volume of water for several days (for example, for 5 days with shungite:water mass ratio of 1:10 and by changing the water once a day). Also, after the washing procedure, chemical analysis of the last washing water should be carried out.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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