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Assessment of the possibility of secondary water pollution during its purification in filtering jugs

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ABSTRACT

The research aimed to evaluate the effectiveness of the overflow filters. In a 2.0 dm^3 filter jug, 5 dm^3 of tap water was filtered daily for one month. The effectiveness of the filters was evaluated based on physic-chemical parameters, that is, the degree of water hardness, pH, chloride content, absorbance, color, and total organic carbon. Taking into account the discussed parameters, it was found that after just two weeks of filter operation, it ceases to fulfill its function. Then, the occurrence of micropollutants in the obtained filtrates was also evaluated. For this purpose, the filtrates obtained for a new filter, a working filter for a month and a filter operating for three months were subjected to a GC-MS chromatographic analysis preceded by solid-phase extraction. The filtrates obtained from working inserts for one month and three months showed, inter alia, the presence of trace amounts of substances included in detergents and personal care products (ethylene brassylate, thymol, tributyl citrate), derivatives of pharmaceutical compounds (methyl ester diclofenac) and industrial admixtures (diisooctyl phthalate, dibutyl phthalate). The source of these substances in the filtered water is the user himself and the products and objects with which the user has a contact in everyday life. In addition, toxicological analysis of the filtration samples was carried out, which showed their non-toxic nature at every stage of the research.

Keywords: Filter jugs; Secondary water pollution; Micro contaminants

1. Introduction

Water constitutes about 60% of an adult's body. Consumption of sufficiently large amounts of water promotes fat burning, body cleaning of toxins, absorption of nutrients and optimal digestive enzymes [1]. In recent years, public awareness about healthy eating, and the quality of water we drink is growing very fast. Therefore, increasing interest is the purification of water secondarily contaminated during transport to the treatment plant to the recipient. One of the methods of drinking water treatment is the use of overflow filters. Their main task is to remove any substances which adversely affect the taste, odor, and color of the water. The aim of the jugs with a water filter is only depriving her of potentially harmful substances and pollutants [2].

Tests of the effectiveness of water filters were conducted by the German consumer magazine "Markt". They showed that the filters could become a habitat of bacteria. The study of "Markt" shows that tap water after filtration contains more germs than before. The reason is that the bacteria accumulate and multiply in dark and moist filter cartridges. Especially if the cartridges are not stored in the refrigerator, but at room temperature. The second reason is too long to use the filter [2,3]. Therefore, we attempt to assess the effectiveness of the filters work overflow.

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While Lanz and Provins [3] surveyed how significant is the need to improve the quality of tap water among the population in the UK. The review was concentrate mainly on the hardness of the water and improving its quality in terms of appearance, taste, and smell. Considering the hardness, they demonstrated that about 14% of households use at least a water softener or buys a product, such as tablets or softening agents descaling. Due to the aesthetic quality of tap water, about 39% of households use tap water filter devices or buy bottled water. Barnaby et al. [2] conducted studies on the effectiveness of arsenic removal from drinking water in the United States using filters to filter jug. They applied for this purpose the most popular companies in the USA, that is, Pur® and Brita® as well as ZeroWater® and Great Value®. They found that only one tested filter, namely ZeroWater®, reduced the arsenic concentration from 1,000 to 2.6 μg/L, well below the permissible 10 μg/L. In addition, the level of all dissolved solids did not significantly affect the ability of the ZeroWater® filter to remove As^{3+} or As^{5+}. On this basis, it was found that the ZeroWater® filtering pot is an effective way of removing arsenic from drinking water [4].

In other studies conducted by Kruszelnicka [4], it was noted that in the case of storage of jug filters and water not by the manufacturers' recommendations, that is, at room temperature, the permissible numbers of mesophilic and psychrophilic bacteria in filtered water were significantly exceeded. This has already been observed in the first week of the use of the new filter cartridge. Psychrophilic bacteria are partially natural microflora in water, and the presence of mesophilic bacteria can sometimes be dangerous because most mesophiles are mesophilic [5].

It was found that there is little research on water purification in filtering jugs. Therefore, the purpose of the presented study to assess the effectiveness of overflow filters. Physicochemical parameters, micro-contamination and toxicity tests were analyzed. The type of micro-contaminants in filtered water was also assessed.

2. Material and methods

2.1. Apparatus

The operation of filter jugs is effortless (Fig. 1). We take off the lid, pour water inside. The container tapers downwards, and the water flows through the filter. After passing through the cartridge, water flows into the empty chamber and does not mix with raw water. The time of the filtration process itself takes about 10 min. In every universal filter cartridge that fits most jugs, the round is composed of two filter layers. The first - activated carbon - according to information from producers, should remove chlorine, phenols, detergents and some heavy metals like lead, mercury and nickel from water. The second part of the filter cartridge is filled with an ion exchange resin that removes magnesium and calcium ions from the water, which is responsible for its hardness, which causes scaling on the vessel walls.

The substrate of the study was tap water filtered through a Brita pit filter. The physicochemical characteristics of the water tested are given in Table 1 in the chapter "Results and discussion".

2.2. Sampling and analytical methods

The research was carried out at the Silesian University of Technology in Gliwice. In a filter pot with a volume of 2.0 and 5.0 L of tap water was filtered daily for one month. Samples for testing were collected every 3 d. Water samples were also analyzed immediately after rinsing the filter and after the first and second filtration from the establishment of a new cartridge and a filter working for three months.

Fig. 1. Filter jug.

Table 1
Physico-chemical characteristics of the water before and after filtration

Sample	pH	Conductivity, μS/cm	UV_{254} 1/cm	Color mg/L	TC mgC/L	Total hardness, $mgCaCO_3/L$	Cl-mg/L
TW	6.6	522	0.010	0.009	35.43	316	70
1 filtration	6.7	358	0.003	0.009	19.17	92	70
2 filtration	6.7	368	0.001	0.009	17.72	108	70
3 filtration	6.6	350	0.003	0.009	16.27	112	70
3 d	6.9	364	0.003	0.008	11.03	168	70
30 d	7.1	394	0.007	0.008	29.62	300	70

TW – tap water

The effectiveness of the filters was evaluated on the basis of measuring physicochemical parameters, that is, the degree of water hardness reduction, pH, chloride content, absorbance, color, and total organic carbon. Specific conductivity and pH were measured with the ELMETRON CPC 505 multifunction device. Total carbon was determined using a Shimadzu carbon analyzer. Absorbance UV_{254} was tested using a Cecil 1000 spectrophotometer. Overall hardness was determined by the edetian method. However, a Dionex ion chromatography was used to measure chlorides. The color was measured using a Merck spectrophotometer. In addition, toxicity studies were conducted using the Microtox® test by Modern Water (Warsaw, Poland). The degree of toxicity was assessed based on the change in light emission by *Vibrio Fischeri* bacteria possessing bioluminescent properties. The experiment was carried out following the Screaming Test procedure of the MicrotoxOmni software.

Then, the occurrence of micropollutants in the obtained filtrates was also evaluated. For this purpose, the filtrates obtained for a new filter, a working filter for a month and a filter operating for three months were subjected to GC-MS chromatographic analysis.

The chromatographic analysis was carried out by the 7890B GC-MS (EI) chromatograph by Perlan Technologies (Warszawa, Poland). The SLB™-5 ms 30 m × 0.25 mm capillary column of 0.25 μm film thickness from Sigma-Aldrich (Poznań, Poland) worked in an oven temperature program adopted from Kudlek [6]. The oven was heated to 80°C (6 min), 5°C/min up to 260°C and 20°C/min up to 300°C (2 min). Helium at a flow rate of 1.1 mL/min was used as the carrier gas. The ion trap temperature was equal to 150°C, while the temperature of the ion source was set at 230°C. The samples were analyzed in the total ion current (TIC) model in the range 50–400 m/z. The identification of the detected compounds was performed by the comparison of their mass spectra with the mass spectra database NIST v17.

The used GC-MS method was adopted from previous studies devoted to the identification of organic micropollutants of an anthropogenic origin in the water environment [6]. The chosen method parameters allow for the determination of a wide range of organic contaminants from the group of pharmaceuticals and personal care products, pesticides, polycyclic aromatic hydrocarbons and industrial additives.

3. Results and discussion

3.1. Physicochemical analysis

The main idea of people who designed jug filters was the ability to improve the taste, smell and color of water, reduce hardness, deprive it of chlorine and mechanical particles coming from water pipes. However, viruses and bacteria are not removed from the water. The use of filters can, therefore, threaten the shortage of minerals essential for health, and even bacterial infection that accumulates on the filter. Table 1 presents the results of measurements of the basic operating parameters of the selected filter. Tap water each time met the quality set by the Ministry of Health [7].

It was found that in all analyzed samples, it remained constant and amounted to 70 mg/L. As you can see, the obtained results do not confirm the information provided by the producers, that is, lowering the concentration of chlorides in filtered water. Also, the color did not change after the filtration process. Its value fluctuated within. 0,008–0,009 Pt mg/L. Similar relationships were observed by Jezierska et al. in the article on methods for assessing the effectiveness of water purification in filtering jugs [8].

Physico-chemical analysis of the filtered water was carried out every 3 d and no significant changes in the pH of the water were found. The table shows the results of the first and last days of filtration. On the other hand, throughout the whole month, the value of pH varied from 6.6 to 7.6.

Analyzing the conductivity value, a decrease of this parameter was observed after the filtration process. In tap water, the conductivity was 522 μS/cm, while after the filtration process it was reduced to 350–399 μS/cm (24%–33%). The obtained results are shown in Fig. 2.

The concentration of calcium and magnesium ions has also changed. On the first day of filter operation, the degree of reduction in overall hardness was 65%. In the next stage of filtration, the reduction of this parameter has significantly decreased. After 5 d, it was 17.7%, while after 24 d only 5%.

It should be noted that the highest efficiency of calcium and magnesium ions removal was observed after the first and second filtration from the insertion of a new cartridge. It can be concluded that the sorption capacity of activated carbon was exhausted at about 50% during this time. The concentration of total hardness during the entire process shown in Fig. 3.

A similar relationship was observed by analyzing absorbance and total carbon concentration. With the consumption of the filter cartridge, decreasing the efficiency of carbon removal was observed. The obtained test results are shown in Figs. 4 and 5.

On the first day of filtration, TC was reduced by 54%, and after 24 d, by only 23%. It can be seen that after two weeks of filter operation, total carbon concentration started to increase from 19.2 to 29.6 mg/L. Similar relationships were obtained when measuring absorbance UV_{254} as an indicator of organic substance content. Based on the obtained results of the absorbance measurement in filtered water, it was noticed that the water quality deteriorated after three weeks.

Taking into account the parameters discussed, it was found that after just two weeks of filter operation, it ceases to fulfill its function. Gizińska et al. [9] observed similar

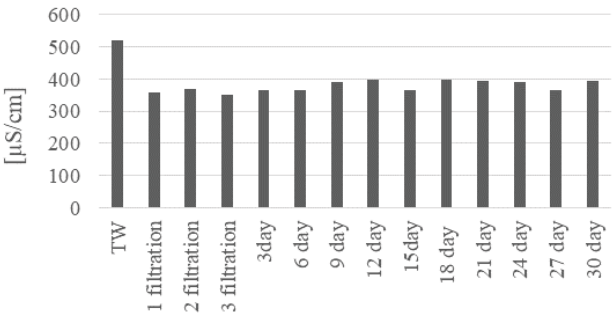

Fig. 2. The conductivity changing during the filtration process.

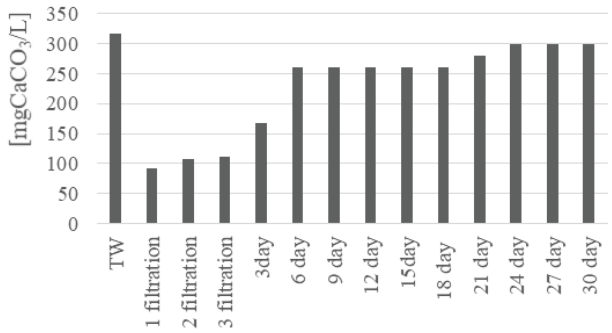

Fig. 3. Change in the value of the general hardness concentration during the filtration process.

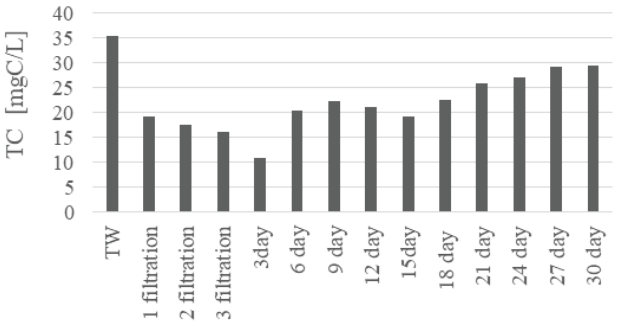

Fig. 4. The dependence of the total carbon concentration during filtration of tap water in the filtering jug.

relations. The authors assessed the effectiveness of jug filters produced by three different manufacturers. The study also showed that the filter pitchers characterized by low retention efficiency of chloride and a slight reduction in total hardness. Gizińska et al. [9] in their work, they also conducted studies on the number of mesophilic microorganisms. In the fourth week of the use of filters, the number of microorganisms developed at the level above 50 CFU-3, which excludes the possibility of more extended use of the filter.

3.2. Chromatographic analysis of the filtrates

The chromatographic analysis of filtrated obtained from the filtration jug after one and three months of cartridge operation indicated the occurrence of several organic compounds (Fig. 6). Only in the case of the filtration carried out by a new filtration jug equipped with a new cartridge no organic compounds were detected (Fig. 6a). The peak signals noted for the filtrate after three months of cartridge operation are significantly higher than those observed for the sample after one month of cartridge use. This phenomenon indicates an increasing concentration of micropollutants in the filtered water. It can be assumed that the micropollutants are continuously adsorbed on the filtration cartridge during the tap water filtration. Before the tap water overflows through the cartridge it comes into contact with the perforated lid of the jug. The lid could be contaminated with micropollutants during direct contact with the skin of the jug user, which can be covered with different personal care

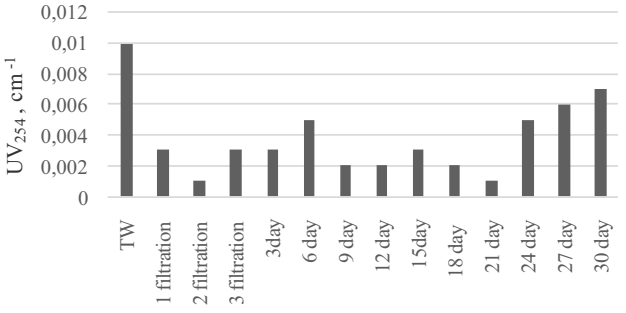

Fig. 5. The dependence of the absorbance value during filtration of tap water in the filtering jug.

products or pharmaceutical compounds. The adsorbed compounds may be subject to desorption and be released into the filtrate.

The second source of micropollutants can be filtered tap water. The literature indicated the presence of several types of compounds in drinking water [10,11]. However, it should be noted that the tap water, which passed through the filtration jug, was tested for the presence of micropollutants during preliminary studies and during the whole duration of the experiment. The test results indicated that tap water was free of compounds identified in the filtrate (Table 2).

The mass spectra of those compounds were compared to the mass spectra database of the United States National Institute of Standards and Technology NIST. This allowed for the identification of single compounds. Table 2 summarized the identification results of compounds, which were characterized by a mass spectra similarity equal to or higher than 70%. This assumption allowed for the match of 22 from 36 possible compounds. Eleven compounds, that is, 2-ethylhexanol, methyl 4-(1,3-dioxolan-2-yl)benzoate, Diethylp, Hedione, 8S,14-cedrandiol, 2,5-Di-tert-butylbenzoquinone, ethylene brassylate, santalcamphor, thymol, butyl citrate and diisooctyl phthalate, belong to the group of personal care product additives. Special attention should be paid to the occurrence of 1-(2,6-dichlorophenyl)-2-indolinone and diclofenac methyl ester, which are the derivatives of the pharmaceutical compound diclofenac.

The jug users decelerated the use of an ointment, which containing this specific non-steroidal anti-inflammatory drug. Therefore it can be assumed that the type of micropollutants detected in the filtrated water strictly depend on the behavior of the users. Another source of micropollutant is tap water, which can also be contaminated by compounds, which passed through the drinking water treatment plant. The literature indicated the presence of different compounds belonging to the group of perfluoroalkyl substances [12,13], pharmaceuticals and personal care products [14–16], pesticides [17,18] and flame retardants [19] in drinking water.

The conducted toxicological analysis pointed on the nontoxic behavior of the collected samples (Table 3). Therefore it can be assumed that the concentration of the detected compounds, which did not exceed 0.1 ng/L, had no negative impact on the water quality. It should be also noted that the compounds get into the water directly from the filtration

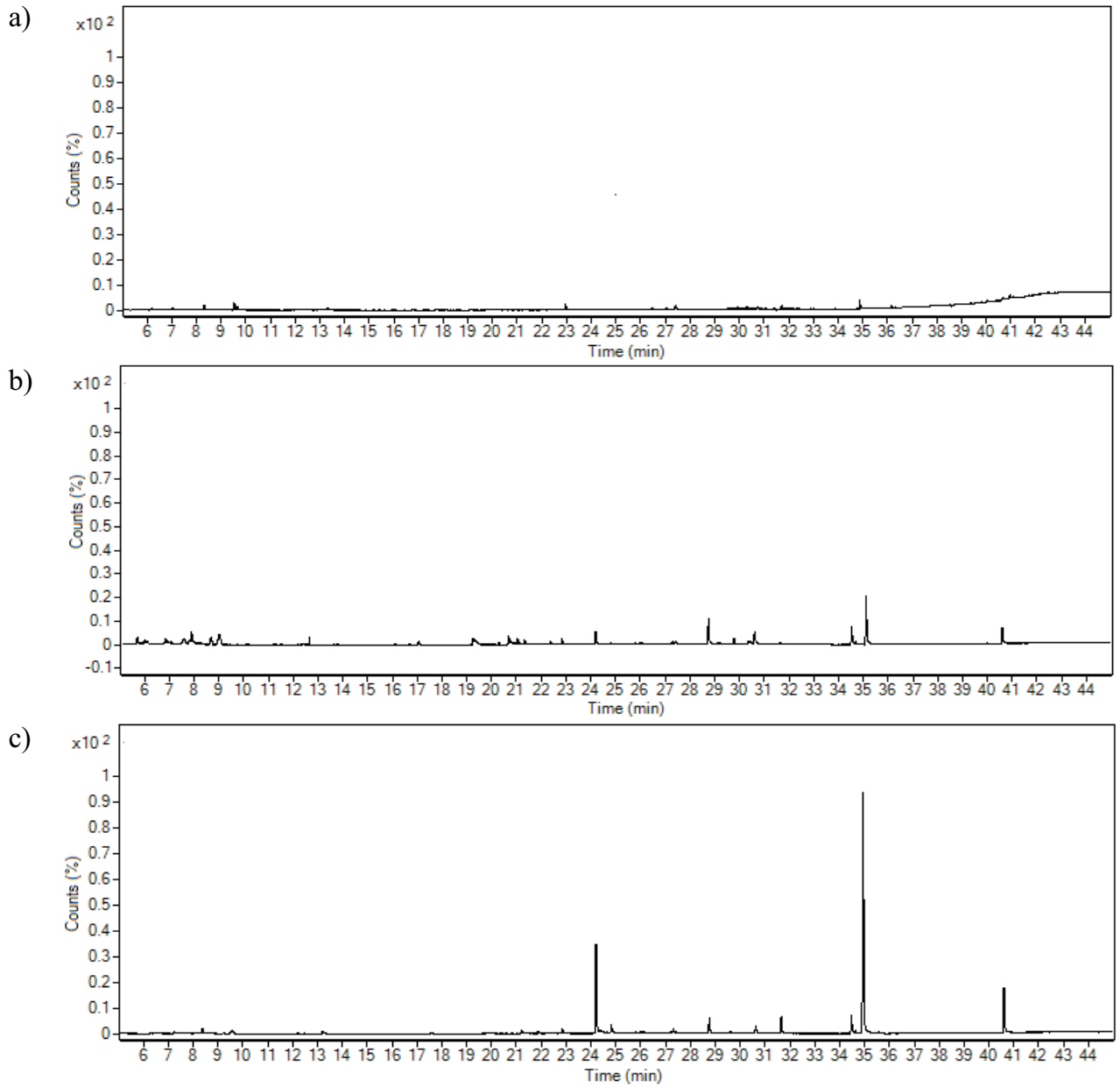

Fig. 6. Chromatograms obtained during the analysis of tap water filtrated trough (a) a new filter and a filter after, (b) one, and (c) three months of operation.

jug user. This indicates that the user has direct contact with a much higher concentration of micropollutants, which can have a negative impact on humans health.

4. Conclusion

The aim of the study was to assess the effectiveness of filter jugs in terms of physicochemical parameters and the occurrence of micro contaminants. Based on the conducted research, the following conclusions can be drawn:

- taking into account the parameters discussed, it was found that after just two weeks of filter operation, it ceases to fulfil its function. The efficiency of removing calcium and magnesium ions decreased on the 18th day of filtration of tap water and was 17%.
- the total carbon concentration after two weeks of testing was 19.2 mg/dm^3, and on the last day of filtration, it was at 29.6 mg/dm^3.
- the chromatographic analysis of water samples collected after one and three months of filter operation showed the occurrence of several organic micropollutants, which gets into the water as a result of the direct contact with the user of the filtration jug.
- the toxicological assessment of filtrates collected during the whole experiment duration indicated no toxicity.

Table 2
Identified compounds

Retention time, min	Compound name	Compound group	Similarity, %
6.27	2-ethylhexanol	Personal care product additive	73
7.23	1-Phenyl-1,2-butanediol	n.d.	72
17.58	2,5-Dihydroxybenzaldehyde	Human metabolite	70
21.21	Modhephene	Propellane product	85
22.25	Methyl 4-(1,3-dioxolan-2-yl)benzoate	Personal care product additive	71
22.85	Diethyl Phthalate	Personal care product additive	86
24.82	Hedione	Personal care product additive	94
25.80	4-Methylsalicylic acid	Dye additive	73
27.31	2,6,10,15-Tetramethylheptadecane	Human metabolite	85
27.43	Tris(1-chloro-2-propyl) phosphate	Industrial additive	81
28.75	Phthalic acid, hept-3-yl isobutyl ester	n.d.	93
29.60	Alachlor	Pesticide	70
29.78	8S,14-cedrandiol	Personal care product additive	70
30.45	2,5-Di-tert-butylbenzoquinone	Personal care product additive	73
30.62	Dibutyl phthalate	Industrial additive	96
31.65	Ethylene brassylate	Personal care product additive	94
32.04	Santalcamphor	Personal care product additive	88
32.48	Thymol	Personal care product additive	76
34.47	1-(2,6-Dichlorophenyl)-2-indolinone	Pharmaceutical compound derivative	89
34.82	Butyl citrate	Personal care product additive	99
35.46	Diclofenac, methyl ester	Pharmaceutical compound derivative	70
40.61	Diisooctyl phthalate	Personal care product additive	98

n.d. – no data

Table 3
Toxicological analysis

Type of sample	Duration of the toxicological test	
	5 min	15 min
Tap water	non toxic	non toxic
Water after the soaking of the filter	non toxic	non toxic
1st filtration	non toxic	non toxic
2nd filtration	non toxic	non toxic
3rd filtration	non toxic	non toxic
day 3	non toxic	non toxic
day 6	non toxic	non toxic
day 9	non toxic	non toxic
day 12	non toxic	non toxic
day 15	non toxic	non toxic
day 18	non toxic	non toxic
day 21	non toxic	non toxic
day 24	non toxic	non toxic
day 27	non toxic	non toxic
day 30	non toxic	non toxic

Calcification according to the guidelines given by Mahugo Santana et al. [20]

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