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HUMAN EXPOSURE TO SILVER RELEASED FROM SILVER-MODIFIED ACTIVATED CARBON APPLIED IN THE NEW TYPE OF JUG FILTER SYSTEMS

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ABSTRACT

Background. A water filtered by jug filter system (JFS) can be applied for the preparation of food products, as well as it can be directly consumed as drinking water. In the European Union, in both above-mentioned cases the quality of water filtered using JFSs has to fulfill the requirements listed in Directive 98/83/EC. However, Directive 98/83/EC sets no parametric value for silver, JFSs are not regulated under this legislative act and additionally, silver-modified activated carbon (applied in such systems) has not been approved by European Food Safety Authority (EFSA). Therefore, the exposure to this metal should be assessed for all JFSs containing filtration cartridges with silver-modified activated carbon, present on the retail market.
Objective. A comprehensive study was conducted in order to examine the effect of JFSs (consisted of filtration oval-cartridges of the new type with silver-modified activated carbon) on the quality of filtered water regarding the released amounts of silver. Silver migration from such type of cartridges has not been examined before. The aim of work was the assessment of exposure to silver released into filtered water from silver-modified activated carbon applied in such types of JFSs.
Material and methods. Silver migration from six brands of JFSs (A–F) was investigated according to British Standard BS 8427:2004 using a validated inductively coupled plasma mass spectrometry method.
Results. The average daily silver concentrations in the composite samples collected on six measurement days for A, B, C, D, E and F JFSs were in the ranges of: 3.95–18.1 μg/l, 4.6–21.7 μg/l, 0.41–8.7 μg/l, 6.9–10.9 μg/l, 3.3–17.1 μg/l and 10.1–20.8 μg/l, respectively. The established grand mean concentrations of released silver from all six oval cartridges were in the range of 2.7–14.3 μg/l. The estimated Hazard Quotient (HQ) indices were in the range of 0.015–0.082
Conclusions. The estimated HQ indices were significantly lower than 1 and therefore no long-term risk for human health could be expected. All the investigated JFSs of the new type meet previously established provisional migration limit for silver from such systems – 25 μg/l.

Keywords: *silver migration, jug filter system, silver-modified activated carbon, human exposure, ICP-MS*

STRESZCZENIE

Wprowadzenie. Woda filtrowana za pomocą systemu filtra dzbankowego (JFS) może być użyta do przyrządzania produktów żywnościowych, jak również do bezpośredniej konsumpcji jako woda przeznaczona do spożycia. W Unii Europejskiej, w obydwu opisanych powyżej przypadkach jej jakość musi spełniać wymagania opisane w Dyrektywie 98/83/EC. Niemniej jednak w Dyrektywie 98/83/EC nie ustalono wartości parametrycznej w przypadku srebra, ww. akt legislacyjny nie obejmuje systemów JFS i dodatkowo węgiel aktywny modyfikowany srebrem (zastosowany w tych systemach) nie uzyskał aprobaty Europejskiego Urzędu ds. Bezpieczeństwa Żywności (EFSA). Dlatego też narażenie na ten metal musi być oszacowane w przypadkach wszystkich systemów JFS obecnych na rynku detalicznym.
Cel pracy. Kompleksowe badania systemów JFS zawierających owalne wkłady filtracyjne nowego typu z węglem aktywnym modyfikowanym srebrem, przeprowadzono w celu określenia wpływu na jakość filtrowanej wody, biorąc pod uwagę wymywane ilości srebra. Migracja srebra z tego typu wkładów filtracyjnych nie była wcześniej badana. Celem pracy było również oszacowanie narażenia na srebro wymywane do filtrowanej wody z węgla aktywnego modyfikowanego srebrem zastosowanego w tego typu systemach JFS.
Materiał i metody. Badania migracji srebra z sześciu rodzajów systemów JFS (A–F) przeprowadzono zgodnie z Normą Brytyjską BS 8427:2004, stosując zwalidowaną metodę opartą na spektrometrii mas z jonizacją w plazmie indukcyjnie sprzężonej.

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Wyniki. Średnie dzienne stężenia srebra w próbkach złożonych uzyskanych podczas sześciu dni pomiarowych w przypadkach systemów JFS - A, B, C, D, E i F, zawarte były odpowiednio w zakresach: 3,95–18,1 µg/l, 4,6–21,7 µg/l, 0,41–8,7 µg/l, 6,9–10,9 µg/l, 3,3–17,1 µg/l i 10,1–20,8 µg/l. Obliczone średnie ogólne stężenia wymywanego srebra ze wszystkich sześciu owalnych wkładów filtracyjnych zawarte były w zakresie 2,7–14,3 µg/l. Wyznaczone indeksy narażenia (HQ) zawierały się w zakresie 0,015–0,082.
Wnioski. Wyznaczone współczynniki HQ były znacząco mniejsze niż 1 i dlatego można stwierdzić, że nawet w przypadku długotrwałego narażenia na srebro wymywane z tego typu systemów nie występuje istotne zagrożenia dla zdrowia ludzi. Migracja srebra w przypadkach wszystkich badanych systemów JFS nowego typu nie przekracza określonego wcześniej tymczasowego limitu migracji srebra z tego rodzaju systemów – 25 µg/l.

Słowa kluczowe: *migracja srebra, system filtra dzbankowego, węgiel aktywny modyfikowany srebrem, narażenie ludzi, ICP-MS*

INTRODUCTION

Silver is not considered as an essential trace element for humans. This metal may be absorbed via the gastrointestinal tract, lungs, mucous membranes, and skin lesions [19]. Silver is accumulated mainly in liver and skin [7, 19]. There were no indications that silver compounds are mutagenic [14] or carcinogenic [8] to humans.

Large doses of this element applied for medical therapies in the form of silver salts have been found to cause argyria. The main symptom of this disease is a permanent discoloration (blue-grey or grey) of the tissues of skin, hair and nails [11]. An oral NOAEL for argyria in humans for a total lifetime intake of 10 g of silver was derived from the available epidemiological and pharmacokinetic knowledge. No newer toxicological data for this metal are available.

Traces of silver can be found in most food products (10–100 µg/kg) while the median daily intake of this metal has been estimated at a level of 7.1 µg [12]. In drinking water that had not been treated with Ag or Ag compounds for disinfection purposes, concentrations of this metal usually do not exceed 5 µg/l [19].

The contribution of drinking water to NOAEL will usually be negligible [21]. Untypical situations may appear when silver or silver salts are used in filtration cartridges of jug filter systems (JFSs) or another point-of-use systems to maintain the bacteriological quality of filtered drinking water. In such cases this metal can migrate into filtered water in amounts that are an essential part of the daily dose of silver ingested by humans.

A JFS of the new type (investigated in this paper) is a freestanding device not connected to a faucet and it consists of: a jug, a hopper, an oval cartridge and a lid. The cartridge is a replaceable container filled with the filtration medium through which unfiltered water passes (gravitationally). The mixture of granular silver-modified activated carbon and weak base ion exchange resins was used as the filtration medium. The main advantage of the application of JFSs for drinking water treatment is the improvement of organoleptic characteristics of filtered water and effective reduction of concentrations of several undesirable chemical substances (e.g. trihalomethanes, pesticides, heavy metals) in a filtrate. The main difference between the oval cartridges of the new type (investigated in this paper) and classic cartridges of the old type (investigated previously) is based on the capacities which are approximately two times higher in the cases of the oval cartridges in comparison to the classic ones. New and classic cartridges are characterized by oval and circular cross sections, respectively.

In this paper a comprehensive study was conducted in order to examine the effect of JFSs (consisted of filtration oval-cartridges of the new type with silver-modified activated carbon) on the quality of filtered test water regarding the released amounts of silver. Silver migration from such type of cartridges has not been examined before. After taking into account the available toxicological data for this metal, the long-term health risk was assessed.

MATERIALS AND METHODS

Instruments and apparatus

An inductively coupled plasma mass spectrometer – XSeries II (Thermo Electron Corporation, UK) was used for the determination of silver in test water. The sample introduction system, the operating conditions and the internal quality control scheme for ICP-MS measurements, as well as validation parameters of the analytical method for silver determination in water by ICP-MS, were described previously in detail [10]. ^{107}Ag isotope was applied for the determination of silver in water samples, while ^{89}Y isotope was used as internal standard.

Reagents, solutions and test water

Calibration solutions were prepared with the use of: deionised water obtained from Simplicity 185 UV system (Millipore SAS, France), UltraPUR concentrated nitric acid (60%; GR; Merck, Germany) and stock solutions - 'Silver ICP Standard 1000 mg/l Ag CertiPUR' and 'Yttrium ICP Standard 1000 mg/l Y CertiPUR' (Merck, Germany). The trueness of the determinations of silver was monitored using certified reference material TMDA-51.3 (Environment Canada, Canada).

The appropriate amounts of test water were prepared on each working day in 20 l HDPE storage tanks (Nalgene, USA), according to British Standard BS 8427:2004 [1, 10].

Investigated cartridges containing silver-modified activated carbon

The tests were conducted for six brands of JFSs with cartridges of the new type (oval cross-section – A, B, C, D, E and F) commercially available on the European, US and Russian markets (bought in Polish retail outlets) – Fig. 1.

Figure 1. Typical filtration cartridges (oval cross-section) applied in the investigated JFSs

Test description

The amounts of silver leached into the test water from JFSs were determined according to British Standard BS 8427:2004 [1] with essential modification described by *Garboś* et al. [10]. Simultaneous examination of two oval cartridges per each type of JFS was performed. The filtering and sampling schedule for JFSs to be tested (up to fixed total filtration capacity – 100 l) was described previously in detail [10, 18]. Samples of both non-filtered test water and composite samples of filtered test water were collected and analysed on the measurement days in the filtration cycle where 5%, 10%, 15%, 25%, 50%, 75% and 100% of the fixed total filtration capacities (100 l) were achieved. In this way, six composite samples were collected on the first, third, fifth, tenth, 15th and 20th working days of the test, including fractions of number of portions of test water filtered: 1–5, 11–15, 21–25, 46–50, 71–75 and 96–100, respectively.

Assessment of human exposure to silver

The daily intake (DI; µg kg^{-1} day^{-1}) of silver was calculated according to the previously described equation [10], after taking into account the established grand mean concentration of silver (GMC_{Ag}; µg/l) for the whole cycle of exploitation of the JFS, the ingestion rate of the filtered water (2 l/day), and body weight (70 kg).

The Hazard Quotient (HQ) for silver was calculated by comparing the estimated DI of this metal with the reference dose (RfD) – 5 µg kg^{-1} day^{-1} [7, 19].

RESULTS AND DISCUSSION

All determined silver concentrations in non-filtered portions of test water were below the quantification limit for Ag (0.014 µg/l).

The results of silver determinations in the composite samples of test water filtered through two independent oval cartridges of each manufacturer (A1/A2, B1/B2, C1/C2, D1/D2, E1/E2 and F1/F2) installed in the corresponding JFSs are given in Table 1. Additionally, average daily silver concentrations (with corresponding standard deviations (SDs)) were calculated for each measurement day (Table 1).

The average daily silver concentrations in the composite samples collected on six measurement days for A, B, C, D, E and F JFSs were in the ranges of: 3.95–18.1 µg/l, 4.6–21.7 µg/l, 0.41–8.7 µg/l, 6.9–10.9 µg/l, 3.3–17.1 µg/l and 10.1–20.8 µg/l, respectively.

The grand mean concentration of silver for the whole cycle of exploitation of the JFS was estimated as the arithmetic mean of all average daily silver concentrations. The established grand mean concentrations of silver for A, B, C, D, E and F JFSs were: 8.2 ± 5.2 µg/l, 9.6 ± 6.5 µg/l, 2.7 ± 3.1 µg/l, 8.4 ± 1.4 µg/l, 7.1 ± 5.4 µg/l and 14.3 ± 3.7 µg/l, respectively.

In 2002 *Garboś* et al. [9] reported for the first time the undesired effect based on leaching silver into water filtered by JFSs containing classic cartridges of the old type (circular cross-section) with silver-modified activated carbon. For two types of cartridges the average concentration of silver in the filtrates was in the range of 19.4–28.7 µg/l. Additionally, the results of tests examining the effectiveness of silver migration process from JFSs of the old type were described in 2003 by the Drinking Water Inspectorate [13], in 2010 by *Święcicka* et al. [18], and in 2012 by *Garboś* et al. [10]. In comparison with the previously conducted study in 2002–2003 [9, 13], the levels of released silver observed during the tests for A–F JFSs were significantly lower. On the other hand, the established grand mean concentrations of silver for A–F JFSs equipped with the new type (oval) of cartridges are comparable with the values obtained for JFSs containing old type (classic - circular) of cartridges that have been tested recently [10, 18].

A part of water filtered by JFSs can be applied for the preparation of food products while the remaining water can be directly consumed as drinking water. In

Table 1. The determined silver concentrations and the estimated average daily silver concentrations in the composite samples achieved for A–F oval filtration cartridges installed in the corresponding JFSs

Percentage of set capacity in the test (%)	Consecutive litres of water applied for the preparation of the composite sample (l)	Silver concentration in the composite sample ± SD* (µg/l)		Average daily silver concentration in the composite samples ±SD (µg/l)	Silver concentration in the composite sample ± SD* (µg/l)		Average daily silver concentration in the composite samples ±SD (µg/l)
		Cartridge A1	Cartridge A2	A JFS	Cartridge B1	Cartridge B2	B JFS
5	1 - 5	8.48 ±0.024	3.26 ±0.023	5.9 ±3.7	9.30 ±0.034	11.5 ±0.023	10.4 ±1.6
15	11 - 15	18.4 ±0.069	17.8 ±0.028	18.1 ±0.42	10.1 ±0.015	11.6 ±0.030	10.9 ±1.1
25	21 - 25	9.58 ±0.0028	9.55 ±0.033	9.57 ±0.021	38.2 ±0.14	5.19 ±0.025	21.7 ±23
50	46 - 50	6.61 ±0.021	6.54 ±0.037	6.58 ±0.049	5.72 ±0.022	4.32 ±0.020	5.0 ±0.99
75	71 - 75	4.81 ±0.015	5.01 ±0.0095	4.9 ±0.14	6.09 ±0.026	4.22 ±0.018	5.2 ±1.3
100	96 - 100	3.88 ±0.039	4.02 ±0.012	3.95 ±0.099	5.70 ±0.0027	3.46 ±0.037	4.6 ±1.6
		Cartridge C1	Cartridge C2	C JFS	Cartridge D1	Cartridge D2	D JFS
5	1 - 5	8.39 ±0.031	9.05 ±0.044	8.7 ±0.47	6.96 ±0.054	7.37 ±0.027	7.2 ±0.29
15	11 - 15	4.13 ±0.049	3.01 ±0.015	3.6 ±0.79	12.3 ±0.023	9.54 ±0.015	10.9 ±2.0
25	21 - 25	2.13 ±0.0076	1.06 ±0.0079	1.6 ±0.76	9.29 ±0.073	6.86 ±0.025	8.1 ±1.7
50	46 - 50	1.17 ±0.011	0.935 ±0.0066	1.1 ±0.17	9.99 ±0.045	7.30 ±0.0057	8.6 ±1.9
75	71 - 75	0.96 ±0.011	0.832 ±0.0024	0.896 ±0.091	10.4 ±0.022	6.80 ±0.027	8.6 ±2.5
100	96 - 100	0.274 ±0.0038	0.554 ±0.0048	0.41 ±0.20	8.3 ±0.15	5.42 ±0.0089	6.9 ±2.0
		Cartridge E1	Cartridge E2	E JFS	Cartridge F1	Cartridge F2	F JFS
5	1 - 5	16.8 ±0.024	17.4 ±0.058	17.1 ±0.42	13.8 ± 0.0052	17.2 ± 0.042	15.5 ±2.4
15	11 - 15	9.65 ± 0.012	9.96 ±0.045	9.8 ±0.22	19.2 ± 0.066	22.4 ± 0.024	20.8 ±2.3
25	21 - 25	4.00 ±0.0012	5.17 ±0.0066	4.6 ±0.83	13.0 ± 0.055	13.7 ± 0.0025	13.4 ±0.49
50	46 - 50	3.78 ±0.0087	3.68 ±0.011	3.73 ±0.071	12.0 ± 0.016	12.2 ± 0.0083	12.1 ±0.14
75	71 - 75	4.24 ±0.013	4.33 ±0.0069	4.29 ±0.064	13.7 ± 0.023	13.4 ± 0.033	13.6 ±0.21
100	96 - 100	3.21 ±0.0046	3.36 ±0.0063	3.3 ±0.11	9.52 ± 0.0017	10.6 ± 0.050	10.1 ±0.76

*Precision of measurements performed by ICP-MS (n=3).

the European Union, in both above-mentioned cases the quality of water filtered using JFSs has to fulfil the requirements listed in Directive 98/83/EC [3], because Regulation (EC) No. 178/2002 [16] defines the quality of water intentionally incorporated into the food after the point of compliance as defined in the Article 6 of Directive 98/83/EC [3]. However, the Annex I (Part B) of this Directive sets no parametric value for silver and additionally JFSs are not regulated under this legislative act. Irrespective of those facts parametric values for silver are defined by the national drinking water legislations in several membership countries of the European Union: Czech Republic (50 µg/l), Denmark (10 µg/l), Poland (10 µg/l) and Slovakia (50 µg/l). Additionally, officially authorized maximum admissible concentrations of silver in drinking water at the levels of 50 µg/l, 100 µg/l and 100 µg/l were established in Russia, Australia and the United States, respectively, although the latest value is listed as optional in Secondary Drinking Water Regulations [20]. Silver concentration level is only controlled in water intended for human consumption in these cases when silver or silver compounds are used for disinfection of drinking water or disinfection of water distribution systems.

In the European Union components of JFSs are regulated by Regulation (EC) No. 1935/2004 [17] on materials and articles intended to come into contact with food and Commission Regulation (EU) No. 10/2011 [2] on plastic materials and articles intended to come into contact with food. All plastic components of JFSs in fact were approved and included in a positive list of the above-mentioned Commission Regulation (EU) No. 10/2011 [2], indicating the specific migration limits related to the appropriate substances. Nevertheless, European Food Safety Authority has not established a specific migration limit for silver released from JFSs containing silver-modified activated carbon. Recently a few questions for written answers [4, 5] concerning potentially harmful effects related to consumption of drinking water filtered by JFSs were directed to the

Table 2. Hazard Quotient (HQ) indices estimated for the whole cycle of exploitation of A–F JFSs

JFS	Grand mean concentration of silver - GMC_{Ag} (µg/l)	Calculated daily intake - DI (µg kg^{-1} day^{-1})	HQ
A	8.2 ± 5.2	0.234	0.047
B	9.6 ± 6.5	0.274	0.055
C	2.7 ± 3.1	0.077	0.015
D	8.4 ± 1.4	0.240	0.048
E	7.1 ± 5.4	0.203	0.041
F	14.3 ± 3.7	0.409	0.082

European Commission. Therefore, the exposure to this metal should be assessed for all JFSs present on the market.

The Hazard Quotient (HQ) indices estimated after taking into account the grand mean concentrations of silver established for the whole cycle of exploitation of A–F JFSs are given in Table 2.

It should be noted that the RfD (which is applied for the estimation of HQ indices) is not a health-derived dose because according to the data published by USEPA it is based on the appearance of argyria (considered as adverse cosmetic effect). Nevertheless, argyria, the critical effect upon which above-mentioned RfD for silver is based, occurs at levels of exposure much lower than those levels connected with other health effects with silver [19]. Thus, no long-term risk for human health can be expected in all cases of the JFSs investigated, because the estimated HQ indices were in the range of 0.015–0.082 (Table 2), being significantly less than 1.

According to the opinion published by European Food Safety Authority [6], for other materials containing silver that were listed in the provisional list published by European Commission [15] of additives used in plastics, a restriction of silver migration into food of 0.05 mg/kg may be allocated. Analogously, the provisional migration limit for silver released from JFSs at the level of 25 μg/l was proposed in previous publication [10]. This value for silver would limit intake to less than 13% of the human NOAEL, under an assumption that each day 2 l of filtered water is consumed containing this metal at the provisional migration limit. All six investigated JFSs investigated (based on the oval cartridges of the new type) meet this requirement.

CONCLUSIONS

1. The established grand mean concentrations of released silver from all oval cartridges installed in corresponding JFSs were in the range of 2.7–14.3 μg/l, being less than the provisional migration limit (25 μg/l) proposed previously.
2. The above-mentioned grand mean concentrations are significantly lower in comparison with the data published in 2002–2003 and therefore, it seems that the producers have modified the technology of the impregnation of activated carbon with silver and lower amounts of this metal were used for this purpose.
3. As silver migrations from the oval cartridges were comparable with the values obtained for the classic cartridges (that have been tested recently) and since capacities of the oval cartridges are approximately two times higher than the classic cartridges, it suggests that lower ratios of the amount of silver-modified activated carbon to the amount of ion exchange resins are presented in the oval cartridges.
4. After taking into account the daily intakes of silver established during the whole cycle of exploitation for six JFSs of the new type (0.077–0.409 μg kg^{-1} day^{-1}) and on the basis of available toxicological data for this metal, no long-term risk for human health could be expected (HQs ≤ 0.082).

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