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Aluminum concentration in drinking water from Moldova territory, Romania

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Abstract Aims of this study were to quantify the levels of Al in tap water from Moldova territory and to compare the obtained value with Romanian Sanitary Norms. Al and other chemical parameters (calcium, magnesium, hardness, sodium, potassium, organic matter, chloride, nitrite, nitrate, microelements) have been determined in the finished drinking waters from Moldova territory (N=80) in 2005-2006 period. Aluminum was determined by molecular absorption spectrometry with Eriocrom Cyanine R (ECR) at a wavelength 535 nm. Method is based on the reaction between Al and ECR, which forms a red dye-lake at approximately pH=6.0. A linear calibration graph was obtained over the range 0.0 to 0.4 mg/L (R^2=0.9984, n=7). The used method gave recoveries from 99.93% to 101.66% for determination of 0.35-0.9 mg Al/L in tap water with satisfactory relative standard deviation values (<5%). Residual Al may vary significantly between different treated waters depending upon conditions, from about 0.003 mg/L to 0.2 mg/L or possibly higher. The obtained analytical data revealed that the Al concentration in drinking water ranged between 0.02 to 0.35 mg/L. 15.9% of all samples had residual concentration of aluminum greater than 0.2mg/L (maximum allowable concentration). The study demonstrates that there is necessary an appropriate drinking water treatment process control in order to ensure the optimum aluminum dose.

Keywords: aluminum, finished drinking water, molecular absorption spectrometry, Eriocrom Cyanine

1. Introduction

Aluminum, the third most abundant element of the Earth's crust, is a non essential toxic metal in human. Aluminum has been considered to be causative agent for various neurological disorders including Alzheimer's Disease (AD) [1, 2]. The World Health Organization estimates that were are 18 million people with AD (4.6%) of the world's population. By the year 2030 it has been estimated that this number will approach 9 million people [3].

Aluminum levels in drinking water vary according to the levels found in the source water and whether aluminum coagulants are used during water treatment.

The purpose of this study, conducted from 2005 to 2006, was:

- to determine the levels of Al in finished drinking water from Moldova territory (Romania);
- to establish the correlations between Al concentration and other chemical parameters in processed surface waters;
- to assess the ingestion of Al with drinking water.

2. Experimental

Thirteen water purification plants were selected from Moldova territory, all of them having as raw material surface water sources as follows: Iaşi (Şorogari, Belceşti, Vlădeni), Vaslui (Delea, Huşi, Negrileşti), Suceava (Dragomirna, Roşu, Colacu, Baia 3), Galaţi (Ţiglina), Botoşani (Bucecea) and Neamţ (Bâtca Doamnei)

Samples were collected from various parts of water treatment process: influent (raw water), coagulation, filtration and finished water (effluent).

In the same time, there were analyzed 60 tap water samples collected from the same locations. Samples were analyzed less than 2 hours after

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sampling. Each sample was run in duplicate and the mean of two successive results at the relative standard deviation not exceeding 5% were accepted as an estimate value.

Aluminum concentration was determined by molecular absorption spectrometry with Eriochrome Cyanine R (ECR). Aluminum ions buffered at pH 6.0, react with ECR dye to produce pink to red complex, in proportion to the concentration of Al. The complex exhibits maximum absorption at 545 nm. Interference from iron and magnesium was eliminated by addition of an inhibitor (ascorbic acid). Standard solution (0.5mgAl/mL) was prepared by using $Al_2(SO_4)_3 \cdot 16\ H_2O$ reagent grade. The following analytical performances have been obtained during the calibration: LOD of 3.5 µg/L (3σ), linear correlation coefficient of 0.9984 (n=7) and range response from 0.0-0.4 mg/L. Also, recovery tests were made, values obtained were from 99.93 to 101.66% of recovery for Al spiked samples.

Other chemical parameters: calcium (Ca), magnesium (Mg), sodium (Na), potassium (K), hardness, organic matters, chloride (Cl), nitrate (NO_3), bicarbonates (HCO_3) according to the Romanian Standards (Law 458/2002) have been determined.

Statistical analysis. Results were expressed as the mean values ± standard deviation ($\overline{x}$ ± SD) using Microcal Excel Windows 2000 and statistical differences between groups were assessed by Student's t test. Values of $p<0.05$ were considered statistically significant. The analytical data were analyzed by Sperman rank correlation analysis.

3. Results and discussions

The use of $Al_2(SO_4)_3$ as coagulation reagent in water treatment process has major and substantial public health benefits. With such use, it is impossible not to have some low level of Al in treated water.

Residual Al content ions in finished water are a function of : (i) the Al levels in the water sources, (ii) the dosing of aluminum - base coagulant, (iii) the pH of water, (iv) temperature, (v) dissolved organic carbon level and (vi) the efficiency of filtration process. It was established that the treatment with $Al_2(SO_4)_3$ removes most of Al associated with particular matter, but introduces a significant amount of dissolved bioavailable Al. Dissolved Al species are complex and can included complexes with natural organic matter, fluoride, phosphate, sulphate.

Temperature, pH and turbidity of the water are important factors in determining Al solubility and consequently residual aluminum [4].

In Table 1 the variation of Al concentrations in different water samples is presented.

The evaluated Al content found in the influent of the filtration unit was: Şorogari 0.251 ± 0.06 mg/L, Negrileşti 0.29 ± 0.21 mg/L, Ţiglina 0.283 ± 0.03 mg/L; after coagulation - filtration stages, the Al level in the effluent was lower than influent.

The variation of the Al concentrations in water samples (raw water, coagulation, filtration and drinking water) collected from the water treatment plant of Suceava district are shown in Fig. 1. The maximum value of Al in finished water was 0.165 ± 0.051 mg/L (Roşu).

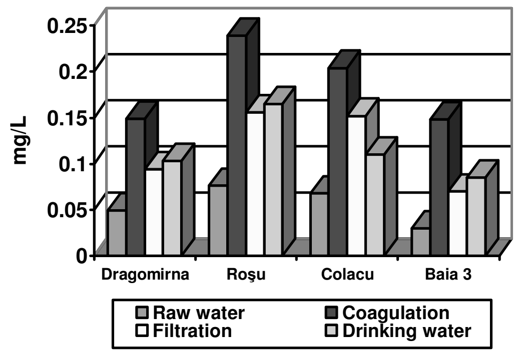

Fig.1. Variation of aluminum concentration in water

As a consequence of aluminum sulphate treatment of surfaces sources, the levels of Al in the treated water were often than those in raw water.

The measurement of Al concentration in the finished water revealed the values for interquartile range (q3-q1) of 0.155 (Delea), 0.075 (Huşi), 0.15 (Şorogari), 0.07 (Vlădeni) (Fig. 2).

15.9% of all samples had residual concentration of aluminum greater than 0.2 mg/L (maximum allowable concentration): Negrileşti 0.288 mg/L, Şorogari 0.35 mg/L, Belceşti 0.23 mg/L.

Concerning the other chemical parameters of studied drinking water, the hardness exceeded 5°G guideline, ranging between 12.1 and 19.4 °G.

Table 1. The variation of aluminum concentration in the studied water samples

Water treatment plant	Source	Aluminum concentration (mg/L)				
		Raw water	Coagulation	Filtration	Finished water	Water to consumers
Şorogari (Iaşi)	Prut	0.0163 ± 0.003	0.251 ± 0.06	0.0165 ± 0.073	0.224 ± 0.098	0.189 ± 0.108
Belceşti (Iaşi)	Tansa Belceşti	0.042 ± 0.007	0.33 ± 0.011	0.21 ± 0.036	0.23 ± 0.046	0.171 ± 0.044
Vlădeni (Iaşi)	Hălceni	0.018 ± 0.001	0.046 ± 0.013	0.023 ± 0.012	0.025 ± 0.008	0.075 ± 0.0405
Delea (Vaslui)	Soleşti	0.044 ± 0.019	0.176 ± 0.149	0.2 ± 0.102	0.17 ± 0.089	0.152 ± 0.07
Huşi (Vaslui)	Prut	0.043 ± 0.031	0.096 ± 0.05	0.08 ± 0.038	0.098 ± 0.089	0.123 ± 0.059
Negrileşti (Vaslui)	Căzăneşti	0.041 ± 0.017	0.29 ± 0.21	0.177 ± 0.134	0.236 ± 0.077	0.185 ± 0.075
Dragomirna (Suceava)	Suceava	0.049 ± 0.007	0.949 ± 0.09	0.094 ± 0.066	0.103 ± 0.059	0.131 ± 0.033
Roşu (Suceava)	Dorna	0.076 ± 0.019	0.239 ± 0.07	0.156 ± 0.021	0.165 ± 0.051	0.037 ± 0.002
Colacu (Suceava)	Moldova	0.068 ± 0.009	0.204 ± 0.08	0.152 ± 0.059	0.117 ± 0.037	0.168 ± 0.091
Baia 3 (Suceava)	Moldova	0.030 ± 0.006	0.148 ± 0.06	0.07 ± 0.039	0.085 ± 0.043	0.105 ± 0.044
Ţiglina (Galaţi)	Dunăre	0.033 ± 0.011	0.283 ± 0.03	0.144 ± 0.011	0.158 ± 0.063	0.238 ± 0.21
Bucecea (Botoşani)	Siret	0.046 ± 0.013	0.27 ± 0.092	0.17 ± 0.068	0.121 ± 0.033	0.129 ± 0.06
Bâtca Doamnei (Neamţ)	Bistriţa	0.0227 ± 0.003	0.226 ± 0.068	0.14 ± 0.04	0.132 ± 0.024	0.113 ± 0.074

Table 2. Sperman rank correlation between factors in drinking water processed from surface water (Şorogari water treatment plant)

	Ca	Mg	Cl	Al	HCO_3	NO_3	Na	K	Org.matter	Hardness
Ca	1									
Mg	-0.093	1								
Cl	-0.075	0.73	1							
Al	0.6122*	0.063	0.226	1						
HCO_3	0.2784	0.548	0.596	0.198	1					
NO_3	-0.279	0.116	-0.323	0.008	-0.0495	1				
Na	-0.433	0.716	0.736*	-0.126	0.374	0.008	1			
K	-0.779	0.624	0.349	-0.503	-0.0088	0.265	0.661*	1		
Org.matter	0.2729	0.018	0.324	0.231	0.2251	-0.425	0.339	-0.369	1	
Hardness	0.586	-0.462	-0.287	0.036	-0.1003	-0.351	-0.541	-0.756	0.4031	1

* $p<0.005$

Table 3. Sperman rank correlation between factors in drinking water processed from surface water (Delea water treatment plant)

	Ca	Mg	Cl	Al	HCO_3	NO_3	Na	K	Org. matter	Hardness
Ca	1									
Mg	-0.6151*	1								
Cl	-0.1278	-0.242	1							
Al	-0.5056	0.191	0.2485	1						
HCO_3	0.2808	-0.427	-0.217	-0.033	1					
NO_3	-0.004	0.187	-0.687	0.082	0.135	1				
Na	-0.1961	0.326	-0.029	0.548	-0.46	0.199	1			
K	-0.8097*	0.359	0.047	0.4398	0.034	-0.13	-0.038	1		
Org. matter	0.0103	-0.001	0.1381	0.0797	0.439	-0.075	0.103	0.2474	1	
Hardness	0.4825	-0.609	0.3403	0.0893	0.651	-0.077	-0.246	-0.172	0.6091**	1

* $p<0.005$ ** $p<0.05$

In generally the level of chlorides, nitrate and sodium, were in accordance with the Romanian Sanitary Norms for drinking water.

Correlation coefficients for some chemical parameters in drinking water are present in Table 2 for Şorogari water treatment plant and in Table 3 for Delea water treatment plant.

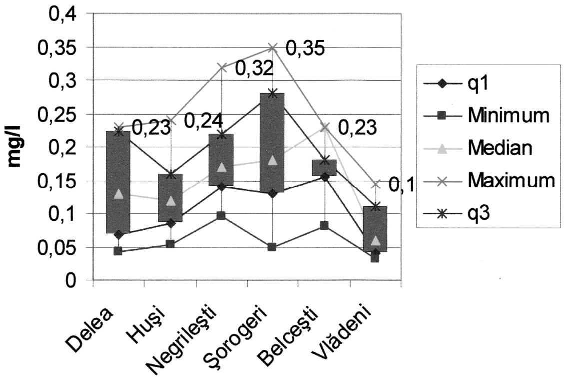

Fig.2. Interquartil range of aluminum in finished water

In the drinking processed surface water (Şorogari) the aluminum has a positive correlation with calcium concentration (r = 0.6122) and a weak negative correlation with other ions (Na, K). There were positive correlation between concentrations of Na-Mg (r = 0.716), Na-Cl (r = 0.736), K-Mg (r=0.624).

Several authors have suggested that a concentration of 0.1 mg Al/L in drinking water appear to provide sufficient protection from AD and other potential neuro toxic effects observed in various epidemiological studies [5, 6]. It was shown that only 1 to 2 percent of the daily intake of Al came from aluminum sulphate treated waters [7, 8].

The calculated ingestion of Al based on the consumption of 2 L of drinking water per day taking in calculation the maximum Al concentration detected in drinking water in different locations are:

- Şorogari 0.44 mg/day corresponding to 0.006 mg/kg bw/day
- Delea 0.342 mg/day corresponding to 0.0048 mg/kg bw/day
- Huşi 0.196 mg/day corresponding to 0.006 mg/kg bw/day
- Colacu 0.11 mg/day corresponding to 0.006 mg/kg bw/day
- Ţiglina 0.144 mg/day corresponding to 0.006 mg/kg bw/day.

Recent studies revealed that Al is not cause of AD, but may contribute to the disease by accelerating a processes initiated by other factors [9].

Nonetheless reduction of aluminum in drinking water is recommended to minimize the population exposure.

4. Conclusions

The obtained analytical data revealed that the Al concentration in drinking water ranged between 0.02 to 0.35 mg/L. 15.9% of all samples had residual concentration of aluminum greater than 0.2mg/L (maximum allowable concentration).

The ingestion of Al based by consumption of 2 L of drinking water was estimated between 0.024 and 0.23 mg/kg bw/day.

The study demonstrates that there is necessary an appropriate drinking water treatment process control in order to ensure the optimum aluminum dose.

5. References

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