Occurrence and Assessment of Selected Chemical Contaminants in Drinking Water from Eastern Romania

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In this study we report on the chemical quality evaluation of drinking water samples collected from Eastern Romania with the main aim on determining the compliance with the official regulation and its influence on the human health for the population living in this area. Therefore, we have collected paired water samples from multiple locations during two seasonal phases (summer and fall) in total being analyzed a number of 80 drinking water samples. By using national standard analytical methodologies, the chemical parameters targeted for analyses included: nitrates, nitrites, ammonium, sulfates, chlorides, trihalometans and organochlorine pesticides. After chemical analysis and data processing 77% among samples were found to be compliant with the Romanian regulation. However, 10% of the samples presented levels above the regulation limits for nitrates and ammonium. Additionally, organochlorine pesticides were measured at levels above the method limits of detection in all analyzed samples while occasionally the pesticide levels were significantly above the regulatory limits. Given the potential health risks on humans health acknowledged for the contaminants measured at high levels in collected samples and combined with the previous literature in the field, this study evidenced the needs for continuous monitoring of drinking water quality at least for the Eastern part of Romania.

Keywords: chemical parameters, drinking water, monitoring, public health

The quality of the drinking water directly affects the human health. Inappropriate use of fertilizers, uncontrolled discharges or leakage from industrial plant, waste water discharges, chemical accidents and disasters can contaminate the drinking water supplies [1-4]. Water is a very important natural resource, vitally significant for the people, but when is polluted it may become the source of several diseases, such as reproductive, cardiovascular and neurological problems or even various cancers [5, 6].

There are international and national legislations to control the water quality consumed by population. Directive 98/83/EC and Normative of World Health Organization [7-9] is one of most recent EU regulation, setting quality standards for drinking water. Drinking water quality regulations established by Directive allows Member States to adapt the monitoring of water quality to local conditions, not only to evaluate the safety for consumption, but at the same time, to detect any toxicological problems occurred and introduce the appropriate measures for restoring and maintaining a good quality of drinking water [8-10]. Although Europe, in general, does not necessarily face problems regarding drinking water quality as it is the case of other parts of the world, there are still water quality problems mainly related to the effectiveness of the water treatment or defects in the distribution networks which may cause risks and adverse health effects to consumers [11].

In this context, the present study aimed on assessing the chemical quality of the drinking water from Eastern Romania during the years 2013–2014, in order to conclude the quality of the drinking water and its impact on the health of the population living in this region. This is achieved by comparing the values of the measured parameters with drinking water guidelines of Romania [6], World Health Organization [9] and the European Union [7].

Experimental part

Materials and methods

Study area and Sample collection

Water samples were collected during the 2013–2014 from ten counties, from the Moldavia region, Eastern Romania (fig. 1). For this study small *Supply Areas* were selected. Supply Area (SA) is a geographical area where drinking water comes from one or more sources within each water quality may be considered approximately uniform. Small SAs are those areas of water supply covered by Directive 98/83 EC which provides a water volume between 10 and 1000 m^3/day.

Samples collection was conducted in two seasonal phases (summer and fall) including two samples of each county. Therefore, a total of 80 water samples were collected during the two years of study.

Analytical methods

The inorganic compounds (nitrite, nitrate, sulphate and ammonium) were analyzed by spectrophotometry using a Agilent 8453 Spectrophotometer (table 1). The chlorides were analyzed using an argentometric method. The organic compounds, trihalomethanes (THMs) and organochlorine pesticides (OCPs) were analyzed by gas chromatography using a Schimazu Gas Chromatograph 2010 equipped with ^{63}Ni selective Electron Capture Detector.

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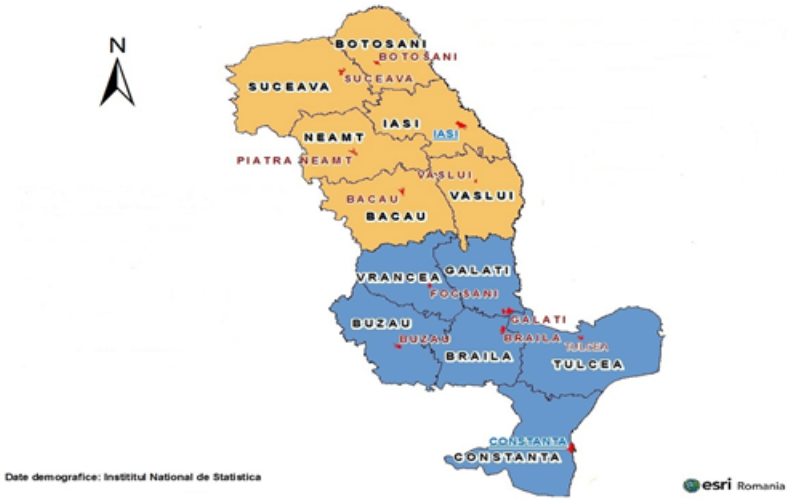

Fig. 1. Map of Eastern Romania with counties included in the monitoring campaigns for this study

Table 1

PARAMETERS AND ANALYTICAL METHODS USED IN ANALYSIS OF WATER SAMPLES

Variable	Method	Reference
Nitrite	Determination of nitrite - Molecular absorption spectrometric method	SR EN 26777:2002
Nitrate	Determination of nitrate. Part 3: Spectrometric method using sulfosalicylic acid	SR ISO 7890-3:2000
Sulphtate	Sulphates content determination	STAS 3069-87
Ammonium	Determination of ammonium. Part 1: Manual spectrometric method	SR ISO 7150-1:2001
Chloride	Determination of cloride. Silver nitrate titration with chromate indicator (Mohr's method)	SR ISO 9297:2001
THMs	Determination of highly volatile halogenated hydrocarbons - Gas-chromatographic methods	SR EN ISO 10301:2003
OCPs	Determination of certain organochlorine insecticides, polychlorinated biphenyls and chlorobenzenes. Gas chromatographic method after liquid-liquid extraction	SR EN ISO 6468:2000

SR – reference standard; ISO - International Organization for Standardization

Statistical analysis

All statistical analyses were performed using XLStat-Pro version 2013.5.01 (Addinsoft, 1995-2013). For data which did not follow a normal distribution (Shapiro-Wilk test, $p > 0.05$) they were log-transformed ($y = \log(x+1)$) and further tested for normality. For data which did not show a normal distribution after log-transformation, non-parametric statistics was applied. For data which followed a normal distribution after log-transforming, parametric statistics was used on the new set of data. Correlations were carried out using parametric Pearson correlations (for normally distributed parameters or log-transformed data). The significance level was set at $\alpha = 0.05$ throughout the study.

Results and discussions

Concentrations of the targeted chemicals in the drinking water samples from Eastern Romania during the period 2013–2014 were summarized in table 2.

Nitrates and nitrites are important indicators of water pollution. In Eastern Romania, the main sources of nitrogen come from numerous natural and man-made sources, including waste waters and agricultural and urban runoff. Nitrates levels ranged from a minimum of 0.17 mg/L in Neamt County (2014) to a maximum of 117.29 mg/L in Botosani County (2014), which value is higher than the maximum admissible concentration. Nine drinking water samples from our study showed nitrates values higher than 50 mg/L - maximum admissible concentration (MAC). The human consumption of water containing such high nitrate levels can cause adverse health effects including methemoglobinemia in infants living in these locations, especially when long term exposure occurs.

Nitrites values were lower than 0.5 mg/L - maximum admissible concentration (MAC) in all locations, with one exception in Botosani county (2013) were 0.54 mg/L nitrite concentration was found.

Many other studies also investigated the levels of nitrogen containing compounds from drinking water samples. Table 3 summaries the levels of some chemical contaminants previously reported for drinking water samples from different countries in comparison with data we report for the present study. Therefore, in Finland (1997), nitrates levels ranged between 0.05–1.19 mg/L these levels being much lower when compared to data reported by our study. Additionally, Finnish drinking water's nitrite level was below the detection limit of 0.01 mg/L [11]. As shown in table 3, the drinking water nitrates levels below the maximum admissible concentration were also previously reported for other countries including Iraq (2010) [12], Turkey (2004) [1], Macedonia (2013) [14], Albania (2013) [15] and Yemen (2013) [16].

D'Alessandro et al. (2012) studied drinking water quality in Italy and determined that Italian water's nitrate levels ranged between 0.05 and 296 mg/L, with a maximum value significantly higher when compared to that of

Table 2

DESCRIPTIVE STATISTICS FOR CHEMICAL PARAMETERS MEASURED IN DRINKING WATER SAMPLES

Compound	2013			2014			LOQ	MAC
	min	max	median	min	Max	median		
Nitrites (mg/L)	< LOQ	**0.54**	< LOQ	< LOQ	0.431	< LOQ	0.01	0.5
Nitrates (mg/L)	0.5	**78.1**	8.575	0.17	**117.29**	4.72	0.05	50
Ammonium (mg/L)	< LOQ	**2.884**	0.032	< LOQ	**1.70**	< LOQ	0.015	0.5
Chlorides (mg/L)	0.5	202.2	27.77	4.26	195.33	30.87	0.01	250
Sulphates (mg/L)	10.61	240.07	72.5	20	201.88	62.09	0.05	250
$CHCl_3$ (µg/L)	< LOQ	69.71	< LOQ	< LOQ	42.75	2.475	1.56	NA
$CHBrCl_2$ (µg/L)	< LOQ	20.74	< LOQ	< LOQ	15.70	1.91	1.38	NA
$CHClBr_2$ (µg/L)	< LOQ	13.30	< LOQ	< LOQ	10.33	2.785	0.54	NA
$CHBr_3$ (µg/L)	< LOQ	13.57	< LOQ	< LOQ	30.50	< LOQ	1.03	NA
Σ THM (µg/L)	1.87	94.24	5.21	2.52	62.13	9.53		100
p,p'-DDD (µg/L)	< LOQ	0.018	0.001	< LOQ	0.005	0.001	0.001	0.1
p,p'-DDE (µg/L)	< LOQ	0.039	0.001	< LOQ	0.079	0.002	0.001	0.1
p,p'-DDT (µg/L)	< LOQ	0.02	0.001	< LOQ	0.025	0.002	0.001	0.1
Σ DDTs (µg/L)	0.0016	0.077	0.003	0.0018	**0.109**	0.005		0.1
α-HCH (µg/L)	< LOQ	0.063	0.001	< LOQ	0.008	< LOQ	0.001	0.1
β-HCH (µg/L)	< LOQ	0.07	< LOQ	< LOQ	0.004	< LOQ	0.001	0.1
δ-HCH (µg/L)	< LOQ	0.001	< LOQ	< LOQ	0.003	< LOQ	0.001	0.1
γ-HCH (µg/L)	< LOQ	0.009	< LOQ	< LOQ	0.013	< LOQ	0.001	0.1
Σ HCHs (µg/L)	0.0015	**0.143**	0.0018	0.0012	0.028	0.0012		0.1
Aldrin (µg/L)	< LOQ	0.013	< LOQ	< LOQ	0.001	< LOQ	0.001	0.03
Dieldrin (µg/L)	< LOQ	**0.168**	< LOQ	< LOQ	**0.051**	< LOQ	0.001	0.03
Hept.epox. (µg/L)	< LOQ	0.009	< LOQ	< LOQ	0.01	< LOQ	0.001	0.03
Heptaclor (µg/L)	< LOQ	0.027	0.001	< LOQ	0.009	0.001	0.001	0.03
γ-clordan (µg/L)	< LOQ	0.04	0.0025	< LOQ	0.04	0.001	0.001	0.1
α-clordan (µg/L)	< LOQ	**0.115**	0.001	< LOQ	0.01	< LOQ	0.001	0.1
En.sulfan I (µg/L)	< LOQ	0.019	0.004	< LOQ	0.004	< LOQ	0.001	0.1
Endrin (µg/L)	< LOQ	0.005	< LOQ	< LOQ	0.001	< LOQ	0.001	0.1
En.sulfan II (µg/L)	< LOQ	0.008	< LOQ	< LOQ	0.007	< LOQ	0.001	0.1
Σ POC (µg/L)	0.007	0.354	0.029	0.006	0.143	0.022	0.001	0.5

LOQ – method quantification limit

MAC – maximum admissible concentration

NA – not available

maximum acceptable limit of 10 mg/L for NO_3^-, recommended for the water to be consumed by infants [17, 18]. It was therefore suggested that such high nitrate levels in drinking water might be responsible for several adverse health effects in children living in these regions especially due to long term exposure.

High concentrations of nitrates were also reported in other countries. Therefore, in northern Greece have been reported nitrate levels of 75 mg/L [10], while in Korea were measured nitrates at 204 mg/L in drinking water samples [19]. Such levels of nitrates in drinking water could be explained mainly due to uncontrolled use of fertilizers in agriculture, the high nitrates concentrations being indeed evidenced in the areas which are characterized by intensive agricultural activity.

The presence of ammonia at higher than geogenic levels is an important indicator of fecal pollution. Taste and odor problems as well as decreased disinfection efficiency are to be expected if drinking-water containing more than 0.2 mg/L of ammonia is chlorinated, as up to 68% of the chlorine may react with the ammonia and become unavailable for disinfection [20, 21]. In the present study, 10% of the analyzed samples presented levels above the regulation limits for ammonium with a maximum level of 2.884 mg/L in Vaslui County (2013).

Chloride and sulphate are major constituents of natural water that are generally good tracers of the sources responsible for increases of water salinity [17].

Chloride is one of the major inorganic anion present in water. The salty taste of drinking water is noticed due to the increased chlorides concentrations. However, since there were not evidenced adverse effects of chlorides on human health, their maximum acceptable levels are generally limited to rather high levels of 250 mg/L [8, 9] in supplies intended for public use. In the present study, chloride levels measured in water samples ranged between 0.2 mg/L and 202.2 mg/L, all of the analyzed samples presented chlorides al levels below the maximum admissible concentration.

The presence of high levels of sulphates in water may contribute to the corrosion of distribution system. Regarding their effect on human health, a high concentration in sulphate, beside the bad taste of the water, it was previously shown that might induce a laxative effect [10]. Regarding the obtained data in the present study, the sulphate levels measured in drinking water from Eastern Romania were generally low, ranging between a minimum of 10.6 mg/L and a maximum of 240 mg/L, all values being below the maximum admissible concentration for this parameter.

Chlorination is the most commonly employed chemical disinfectant in drinking water treatment nowadays in order

Table 3

DATA REPORTED FOR WATER SAMPLES COLLECTED FROM EASTERN ROMANIA (2013-2014) COMPARED TO OTHER STUDIES IN THE FIELD

Location/ Year	NO_3^- mg/L)	NO_2^-/mg/L	NH_4^+/mg/L	Cl^-/mg/L	SO_4^{2-}/mg/L	ΣTHM/µg/L	ΣPOC/µg/L	Ref.
	Mean/Range	Mean/Range	Mean/Range	Mean/Range	Mean/Range	Mean/Range	Mean/Range	
Finland/ 1997	0.05–1.19	≤0.01	-	-	-	-	-	11
Greece/ 1998	-	-	-	-	-	5-106	-	25
Taiwan/ 2002	-	-	-	-	-	8.3±0.5	-	26
Turkey/ 2004	-	-	-	-	-	60	-	22
Italy/ 2004–2005	0.05-296	-	-	5.53-130.2	5.03-516	-	-	17
Greece/ 2007	12.3	≤0.01-0.34	Max. 5.7	Max 289.7	Max 750	-	-	10
Pakistan/ 2007	-	-	-	-	-	-	0.001-0.080	36
Korea/2008-2011	12.4–204	-	Max. 0.21	Max. 599	2– 185	-	-	19
Irak/ 2010	≤1-7	≤0.025-1.49	-	49-250	157-500	≤1-116	-	12
China/ 2011	-	-	-	-	-	-	0.003-0.064	37
Italy/ 2011	≤0.1–11	-	-	5-102	7-60	-	-	18
Pakistan/ 2015	-	-	-	-	-	103.5	-	28
Turkey/ 2013	4.79±4.20 0.28–16.2	≤0.08	-	-	-	6.63±5.14 2.34-19.3	≤LOQ-0.042	1
Macedonia /2013	3.34 0.2-22.5	-	-	6.16±7.2 0.5-25.8	-	-	-	14
Albania/ 2013	1.87–2.56	0.001- 0.003	0.001-0.05	-	-	-	-	15
Yemen/ 2013	9–28	0.01 -0.092	0.06-0.116	14-58	19-45	-	-	16
India/ 2013	-	-	-	-	-	274-511	-	26
Romania/ 2013	17.8 0.5–78.1	0.033 0.003-0.54	0.239 0.01-2.88	45.35 0.5-202.2	91.71 10.6-240	17.88 1.87-94.24	0.0451 0.007-0.354	Present study
Romania/ 2014	19.10 0.17–117.2	0.030 ≤0.01-0.431	0.077 ≤0.01-1.70	52.66 4.2-195.3	82.55 20-201.8	16.73 2.52-62.34	0.038 0.006-0.142	Present study

to protect the public from waterborne diseases and to control or prevent the regrowth of microorganism and biofilm formation in the distribution system [22, 23]. Chlorine is a relatively unstable chemical compound which reacts with natural organic matter, commonly measured in terms of total organic carbon and inorganic substances, such as bromide ion in water, causing the formation of various chlorination by-products (DBPs) [1, 24]. Due to concerns on the humans health effects reported for organic by-products of chlorination, the use of alternative disinfectants such as chloramines, ozone, and chlorine dioxide as primary or secondary (distribution system) disinfection agents is increasing in the late years worldwide. However, each of these alternatives has also been shown to form its own set of DBPs. A variety of chlorinated DBPs, including trihalomethanes (THMs), haloacetonitriles and haloacetic acids were reported to be commonly formed [25]. The frequently formed THM compounds are the following: trichloromethane (chloroform), bromodichloromethane, dibromochloromethane and tribromomethane (bromoform). These THM species are considered to be toxic, accumulative and persistent in the environment [1]. Several epidemiological studies assessed the carcinogenic potential of these products and negative effects on human health have been proven [26, 27].

In the present study, THM levels measured in the water samples ranged between a minimum of 1.87 µg/L and a maximum of 94.24 µg/L, all of the obtained results being below the maximum admissible concentration recommended by the national and international legislation, which is 100 µg/L [7-9]. Thus, given the toxicity acknowledged for these chemicals, our results did not show an indication of the existence of pollution hazards in the studied area. When compared to the previous literature in this field (Table 3), the measured THM concentrations in Romanian drinking water samples included in this study they were much lower than the reported levels for samples collected from Iraq (2010) [12], India (2013) [26], Turkey (2004) [22] or Pakistan (2015) [28].

In this study we also assessed the organochlorine pesticide contamination in drinking water samples from Eastern Romania. Due to their physico-chemical properties and environmental behavior, out of persistent organic pollutants, organochlorine pesticides are one of the most intensively studied classes of compounds. Humans are exposed to mixtures of such contaminants which individually might have a heterogeneous behavior in the

environment, while they can possibly have an additive effect in terms of toxicity. They were proved to be highly lipophilic substances that persist in the environment, bioaccumulate through the food chain and pose a risk of causing adverse human health effects including immunotoxicity, reproductive effects, endocrine disruption or carcinogenicity [29]. Previous literature in the field of chlorinated pesticides in samples from Eastern Romania evidenced relatively high levels of these contaminants in food, soil and biological human samples, such levels being later confirmed by systematic investigations of human serum samples collected from clinical healthy population in Eastern Romanian [30, 31], human hair sampled from adolescents [32], followed by estimation of the human exposure sources of Romanian population to several organic contaminants through food consumption or indoor dust ingestion [33, 34]. Therefore, although they are lipophilic compounds, their presence in water samples should be limited while measurable levels of such compounds in this matrix might contribute to the total burden of organic pollutants in humans living in this area.

The European [7] and Romanian legislation [8] sets maximum allowable concentrations for pesticides in drinking water at 0.1 μg/L for each active ingredient and 0.5 μg/L for the sum of all chlorinated pesticides. For Aldrin, Dieldrin, Heptachlor epoxide and Heptachlor, due to their high toxicity and persistence in the environment, the legal limit is set at 0.3μg/L of water. Therefore, the results obtained in this study evidenced that all water samples contained measurable levels of various chlorinated pesticides, while the sum of targeted organochlorine pesticides was in the range of 0.006 to 0.354 μg/L, these levels being lower when compared to the maximum acceptable limits for drinking water. Even if the use of organochlorine pesticides has been restricted or even banned throughout the EC, nevertheless, they are still present in drinking water. Although the use of lindane (γ-HCH) and DDT has been banned in Romania in the late '80s, throughout this study we evidenced the measurable levels of lindane, *p,p'*-DDD, *p,p'*-DDE and *p,p'*-DDT in 41.2, 78.7, 52.5, and 55% of the analyzed samples respectively. Additionally, two locations, namely Botosani and Iasi county, presented higher concentrations of these compounds when compared to the maximum recommended levels: in Botosani county SHCHs was measured at 0.143 μg/L, while in Iasi county, SDDTs was measured at 0.109 μg/L. Other pesticide formulations were also measured above the recommended national limits in some locations included in our study, as follows: α-chlordane was measured at 0.115 μg/L in Tulcea county, dieldrin measured at 0.051 μg/L and at 0.168 μg/L in Iasi and Tulcea county respectively (table 2).

As previously reported, two HCH formulations were used in Romania in the past: lindane, containing only the γ-HCH isomer and technical lindane which consisted in a mixture of HCH isomers in the following proportions: 60–70% of α-HCH, 5–12% of β-HCH and 10–12% of γ-HCH [35]. The measurable levels for α-HCH in water samples suggest the use of technical lindane in the past, while occasionally higher γ-HCH levels suggest the past use of pure lindane formulation. However, the low α-HCH/ γ-HCH ratio obtained for the water samples included in this study combined high β-HCH/(α+ γ)-HCH ratio and β-HCH being the dominant HCH isomer for the big majority of investigated samples suggest that the HCHs are present in collected water samples due to the historical use of lindane.

The technical mixture of DDT used in Romania contains 65–80% of *p,p2* -DDT, but also 15–21% of *o,p2* -DDT and up to 4% *p,p2* -DDD [35]. Among DDTs, the dominant compound measured in collected samples was *p,p2* -DDE, the principal metabolite of *p,p2* -DDT which indicates the past use of such pesticide formulation. However, the measurable levels of *p,p2* -DDT in collected water samples is contrasting given the lipophilic nature of this contaminant. Previous research in the field of halogenated pesticides from biological samples collected from the same geographical area as the water samples included in this study [30-34] evidenced the high levels of the DDTs in general and of *p,p2* -DDT in particular in all matrices including human serum and hair, food and indoor dust samples. As a consequence, although *p,p2* -DDT water levels reported in this study are within maximum acceptable legal limits, combined with previous published data from Romanian samples in this field suggests the existence of DDT sources in the investigated area. Supplementary, the data reported in this study also suggests the existence of fresh input sources of heptachlors, aldrins and endrins in addition to the historical residues.

The literature on chlorinated pesticides and other contaminants from other European countries is rather scarce and this might be attributed to the fact that water supply networks are generally used for other purpose than drinking. As a consequence the scientific interest for targeting lipophilic analytes in such samples especially from Western European countries decreased given the low risk for human exposure to these contaminants. It was previously reported that pesticides levels in biological samples such as human serum from Western European locations is significantly low when compared to Eastern European countries [31] and therefore when addressing human exposure to such contaminants is generally oriented to samples with a higher probability to contain these bioaccumulative contaminants, such as food.

When referring to previous literature in this filed on samples collected from other locations, Usman et al. (2014) described an extensive study from Pakistan on chlorinated pesticides and their metabolites by monitoring 79 samples of cattle drinking water in Karachi [36]. Among those, 8% of the samples were contaminated at measurable levels with HCHs and 3% with DDTs. According to author's suggestions, HCH contamination was attributed to industrial activities while those of *p,p'*-DDT and *p,p'*-DDE were attributed to the surrounding persistent pesticide contaminated soils [36]. Another study performed by Wu et al. (2014) included a total of 27 water samples from the Taihu Lake region, China which were monitored for 14 organochlorine pesticides in order to identify their possible sources, and estimate human health risk of the drinking water samples [37]. Therefore, it was shown that, with the exception of *p,p2* -DDD and *p,p2* -DDT, all other targeted chlorinated pesticides occurred at measurable levels with high detection frequencies in all of the water samples of Taihu Lake region. Additionally, it was evidenced that the dominant chlorinated pesticides in investigated samples were DDTs and HCHs accounting for up to 44.2% of the total organohalogenated pesticides targeted for analysis.

Conclusions

This study shows that although strict regulations exist on monitoring the drinking water quality in Romania, still a significant number of samples could be found to not comply with the recommended maximum concentrations for some chemical parameters including nitrates, ammonium and some organochlorine pesticides. Moreover, even if most of the samples complied with the legal regulations from the organic contaminants point of view, all

investigated water samples presented measurable levels of pesticides officially banned in Romania many years ago. The findings of our study is in agreement with previously published work in the field or persistent organic pollutants monitored from human biological samples showing that population from Eastern Romania are highly exposed to such compounds. Given that water supplies included in this study are generally used by population for direct water consumption, combined with the bioaccumulative properties of some investigated contaminants (chlorinated pesticides) this might pose a risk for humans living in Eastern Romania.

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