20

Evaluation of Drinking Water Quality in Three Municipalities of Romania: The Influence of Municipal and Customer's Distribution Systems Concerning Trace Metals

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1. Introduction

The purpose of every water utility is to provide consumers with drinking water that presents no risk to public health. Safe drinking water is generally obtained by complying with specific water quality standards such as European Union Drinking Water Directive (Council Directive 98/83/EC, 1998).

Corrosion can affect public health, public acceptance of a water supply and the cost of providing safe drinking water. The release of corrosion by-products of household plumbing systems can be a significant source of trace metals found in tap waters (Agatenos & Okato, 2008). Pollution with metals may originate mainly from old and poor quality distribution networks and piping systems.

When drinking water is distributed through pipelines, biofilms will grow on the inner surface of the pipes and soft deposits (organic and inorganic matter) and several metals will accumulate to the pipelines (Lehtola & al, 2004a). Discoloration of drinking water is one of the main reasons customers complain to their water company. An elevated concentration of iron or increased turbidity, affect taste, odor and color in drinking water. Unlined iron pipes in drinking water distribution networks develop extensive internal corrosion scales as the time of use increases. These corrosion scale deposits reduce the hydraulic capacity of the pipes and more energy is required to deliver water at a desire flow rate (Sarin et al., 2004).

The heaviest corrosion was observed mainly in steel and cast iron pipes (Nawrocki et al., 2010). Corrosion of cast iron and iron products is ofen seen as the main source for discolouration of drinking water, iron higher content and may can also induce a chemical decay of the residual chlorine (Lehtola et al., 2004b; Sarin et al., 2004).

High contents of Cu can be released in drinking water as an effect of using the copper or copper alloys pipes for cold-water domestic installation. In copper pipes, the concentration of copper in water incresed with increasing stagnation time (Lytle & Schrek, 2000; Merkel et al., 2002). Merkel et al found that during stagnation, the concentration of copper reached its maximum after 10 hours, and then it started to decline. Water stagnation for more than 4 hours significantly increased the copper concentration in water (Lehtola et al., 2007; Lytle & Nadagouda, 2010). In Romanian legislation, the maximum admissible value for Cu in drinking water (100 μg/L) is lower than the limit imposed by EU Drinking Water Directive (2,000 μg/L).

The major source of lead in drinking water was identified to be plumbing materials. Lead pipes, lead – based solder, brass fittings and plumbing fixtures such as pipe's jointing faucets are known to be dominant lead sources in public water supply systems (Kimbrough, 2001; Lasheen et al., 2008).

Lead pipes were replaced with other types of pipes such as polymer materials like polyvinyl chloride (PVC), polyethylene (PE) and polypropylene (PP). Plastic pipes currently make up about 54% of all pipes installed worldwide. PVC makes up 62% of this demand and PE in its various forms about 33.5% (Raynand, 2004).

PVC polymer is mixed with a number of additives including stabilizers in order to provide the range of properties needed in the final products. Stabilizers are often composed of salts of metals like lead and cadmium (Kim, 2001). Unlike PVC, other plastics including PE and PP do not require metallic heat stabilizers. In addition, galvanized iron pipe can release significant amounts of lead into drinking water, as the zinc coating contains about 1% lead impurities (AWWA, 1996).

Nickel can be leached from the internal surface of some components of domestic installations such as: Cr/Ni plated devices, alloys containing nickel (Gari & Kozicek, 2008).

In the process for drinking water production, usually, the raw water is treated with aluminium sulphate as coagulation agent, for this reason is important to control aluminium. The role of Aluminium in Alzheimer illness is not well known but is certainty known that this metal has a toxic effect on the nervous system (Flaten, 2001). Aluminum has been shown to play a causal role in dialysis encephalopathy (Alfrey et al., 1976) and epidemiological studies suggest a possible link between exposure to this metal and a higher prevalence of AD (Flaten, 2001; Becaria et al., 2006). This association is dependent on the duration of Al exposure and only becomes significant if an individual has resided in an area with high Al in drinking water (>100 μg/L) for several years (Becaria et al, 2006).

Drinking water produced in water plants almost invariably fulfils the water quality requirements set in European Union Drinking Water Directive (Council Directive 98/83/EC, 1998). However, the DWD requires that water quality should also meet the requirements at the consumer's tap.

In some European countries such as Romania or Germany, the water distributors must ensure that microbial and chemically clean water reaches water meters. After that, the owner of the building is responsible for the water quality. Up to the water meter, the drinking water quality is very good, but the drinking water collected from the customer tap could have a lower microbial and chemical quality than the water produced and distributed. (Volker at al., 2010).

In most European countries, drinking water quality is not monitored routinely at household level but rather directly in the distribution system, as waterworks and authorities have limited access to private homes, as well as limited control over household plumbing and operation.

Significant levels of trace metals could be detected after stagnation of the water in distribution system, especially during night – time (Haider et al., 2002; Vasile et al., 2009, 2011; Zietz et al., 2003, 2007). All these studies reported increased concentration of lead, cadmium, copper, iron and nickel after stagnation in household tap water in Austria, Germany and Romania.

2. Aim

Tap water from the municipal supply system is the source of drinking water for majority of consumers in Romania. The main sources of drinking water in Romania include rivers (about 60%), drillings and much less, lakes.

According to Romanian legislation, the last segments checked by the Water Companies are branch pipe and water meter. Less than 0.1% of domestic network of customer is included in monitoring plan of drinking water; usually, the tap water is controlled only at customer request or complaint.

The aim of the study was to identify issues that may affect public health and the risk prevalence of relevant metals in in-building installation systems, in three important municipalities from Romania. Thus, it is possible to take measures to increase security of water systems, replacement of the pipelines at risk of metal corrosion, improve drinking water quality and protect human health against adverse effects caused by contamination of drinking water.

In order to get an overview of the overall current contamination levels of drinking water at the point of consumption were collected and analysed more than 600 samples from cold line-pipe with three different sampling procedures (first draw, fully flush and randome daytime).

A monitoring plan was developed and the content of metals (Al, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Se, Sb and Zn) was analyzed from drinking water samples collected directly from consumers and from the municipal distribution network.

3. Methodology of the study

3.1 Cases under study – Municipalities of Bucharest, Timisoara and Tg. Mures

A large distribution system delivering drinking water to about 1.7 million citizens, supplied by three water treatment plants and two different surface water sources was investigated. Bucharest, Romania's capital, is the largest city, the industrial and commercial center of the country, the sixth city of UE population. The Capital is located in south – east part of Romania, in Muntenia Region, in the Romanian Plain. Bucharest has the same administrative level as a county and is divided into six districts.

Three Drinking Water Plants, located outside the city perimeter, supply the city. Arges River, raw water source for Crivina and Rosu Water Plants, is affected by anthropogenic pollution; the river collects wastewater discharges from industrial areas located downstream near Curtea de Arges and Pitesti cities. In case of accidental pollution of both rivers uses as raw water (Arges and Dambovita Rivers), the operator, APA NOVA Bucharest, has in place necessary means to eliminate the pollutants.

Second city selected was Timisoara, Timis County capital, situated in West Part of Romania, in historical region of Banat, the third largest city in Romania. The most important raw water source in collected from Bega River, which is clogged and the water is cloudy for most of the time. The water and sewerage operator, which provide drinking water in Timisoara city is AQUATIM Company.

Targu Mures is the sixteenth largest city in Romania and the sixth in Transylvania, located in central – northern Romania and represent Mures County municipality residence. As only source of raw water is used Mures River. The river collects discharges from several locations upstream of Tg. Mures, most important being Reghin and Toplita cities.

In Tg. Mures municipality, AQUASERV Company is the producer and distributor of drinking water.

In table 1 are presented characteristics of the utilities under study, such as population served, source of raw water, supplied flow rate, water plants, and most important, treatment processes.

	BUCHAREST			**TIMISOARA**			**TARGU MURES**
Population served	1725,000 inhabitants (December 2009), 81% from Bucharest population			330,000 inhabitants (December 2010), 99% from Timisoara population			143,000 inhabitants (December 2010), 95,25% from Tg. Mures population
Source of raw water	*Arges River*		Dambovita River	Bega River	Groundwater (63 drilling points)		Mures River
Water Plants	*Crivina*	*Rosu*	Arcuda	Bega	Urseni	Ronat	Tg. Mures
Supplied flow rate (m^3/day)	259,200	400,000	610,000	69,676	26,131	1,311	46,000
Treatment processes	*pre-ozonation*→coagulation (aluminum sulphates) → flocculation → decantation → inter-ozonation**→ sand filtration → disinfection with chlorine gas* *, ** only in Crivina Water Plant			*Coagulation (aluminum sulphates, sodium aluminates, aluminum polychloride) → flocculation → decantation → sand filtration → disinfection with chlorine gas*			*Pre-oxidation with $KMnO_4$ → pre-decantation → coagulation (aluminum poly hydroxyl chloride) → flocculation → decantation → sand filtration → ozone treatment → adsorption on granular active carbon → disinfection with chlorine gas*

Table 1. General characteristics of the utilities under study

3.2 Sampling

3.2.1 Sampling techniques

In order to obtain a large database, the samples were collected from customer's cold line-pipe with three different sampling techniques: first draw sampling (from kitchen); fully flushed sampling procedure after flushing five minutes same tap; random daytime procedure (within office hour, without previous flushing of the tap).

The definition of a *first draw sample* is a sample that is taken first in the morning before the tap in the premise has been used for other purposes. During the stagnation period no water should be drawn from any outlet within the property (this includes flushing of toilets). If any water is drawn during the stagnation period the result will be invalid. It is common practice for such samples to be taken by consumers. There is no control over the quality of the samples. When the sample is taken, the tap should be fully opened or as open as possible without losing sample (WHO & COST Action 637, 2009; Karavoltos et al., 2008).

The definition of a *fully flushed sample* is a sample that is taken after prolonged flushing of the tap in a premise in such way that stagnation of water in the domestic distribution system does not influence the quality of tap water. In practice a sample is taken after flushing at least three plumbing volumes. In case that the temperature of the water from the distribution network is cooler than the ambient temperature, an alternative method is monitoring the temperature of the water during flushing until it stabilises (WHO & COST Action 637, 2009; Karavoltos et al., 2008).

The definition of a *random daytime sample* is a sample that is taken at a random time of a working day directly from the tap in a property without previous flushing. When the sample is taken the tap should be fully opened or as open as possible without losing sample (WHO & COST Action 637, 2009; Karavoltos et al., 2008).

Each type of sampling technique provides different informations about influence of materials used in both distribution systems (municipal and domestic) to the tap water quality. Thus, the results obtained with fully flushed procedure indicates the influence of municipal distribution system to the tap water supplied by the Operators.

The first draw results shows in principal the influence of materials used in domestic distribution system.

In Europe, random daytime (RDT) sampling (1^{st} liter taken during office hours, without fixed stagnation) and sampling after 30 minutes of stagnation (30 Ms) (1^{st} and 2^{nd} liter) were identified as the best approaches for estimating exposure and detecting homes with elevated lead concentration in tap water (Deshommes, 2010; Hayes, 2009; Hayes et al., 2010).

3.2.2 Sampling points

In order to control drinking water quality, in the study were collected samples from producer via distribution system to consumer's tap, from different points such as:

- Water Plants - drinking water produced by the Water and Sewerage Operators;
- monitoring points of the Operators, situated in different locations in the municipalities, such as elementary and high schools, kindergartens, markets, fountains, public institutions, pumping stations (fully flushed samples);

- selected customers of the Operators with the residence in different districts of the municipalities (first draw and fully flushed samples);
- medical centres, pharmacies, schools, private companies, public institutions, food markets, fast foods, restaurants situated in old buildings from the center of the cities – (random daytime samples).

In the study were collected and analyzed also surface and groundwater used as raw water by the Operators.

The drinking water samples from Water Plants and monitoring points of the Operators were collected by the specialists of the NR&DI ECOIND and the Operators, in a common programme. The customer’s tap water were collected after an adequate instruction by the customers, which were workers from NR&DI ECOIND in Bucharest and Timisoara, AQUATIM (Timisoara) and AQUASERV (Targu Mures) Companies.

The random daytime points were selected and drinking waters were collected by the scientists from NR&DI ECOIND.

In Timisoara and Tg. Mures were collected samples in two campaigns, one in winter season, the other one in summer time. In addition, in summer, from old, historical buildings situated in the center of the cities, were collected drinking water samples with Random Daytime procedure, in order to control lead concentration in tap water.

The technique used for sampling of drinking water from Operator’s monitoring points was fully flushed procedure.

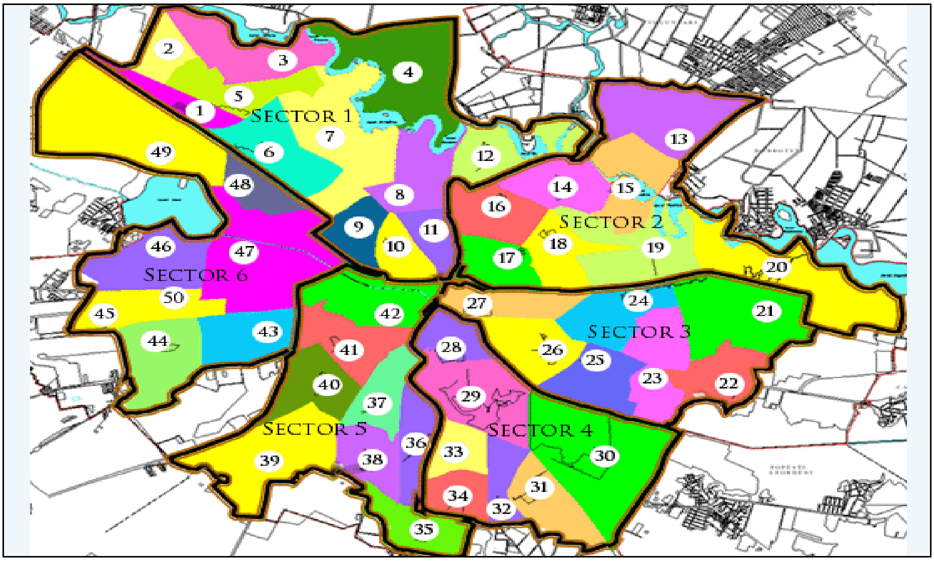
In table 2 are presented data regarding number of samples collected in the study for each municipality and also the date when the samples were collected.

Municipality	Samples									
	Water Plant		Operator Monitoring points		Customer points					
	No	Date	No	Date	First Draw		Fully Flush		Random Daytime	
					No	Date	No	Date	No	Date
Bucharest	3	22.06.09	23	23.06.09	71	22-24.06.09	71	22-24.06.09	-	-
Timisoara	3	16.02.10	15	16 - 18.02.10	30	17.02.10	30	17.02.10	-	-
	3	17.02.10								
	3	18.02.10								
	3	23.06.10	15	23 – 24.06.10	30	24.06.10	30	24.06.10	32	24.06.10
	3	24.06.10								
Tg. Mures	4	16 - 18.02.11	30	16.02.11	18	16.02.11	18	16.02.11	-	-
			30	17.02.11	17	17.02.11	17	17.02.11		
	1	30.06.11	30	30.06.11	17	31.06.11	17	31.06.11	45	30.06.11

Table 2. Number of samples collected in the study

3.2.3 Maps of sampling points distributed in the cities

The map of supply zones with their monitoring points in the water distribution system of the Bucharest Water and Sewerage Operator, APA NOVA, is presented in figure 1. In the monitoring plan were included eighteen points on the treatment flows, in hydro technical nodes, at the six pumping stations (Drumul Taberei, Preciziei, Grivita, Grozavesti, Nord, Sud) and in the distribution network.



Legend: Points: 1-5, 9, 10, 12-15, 19, 24-26, 28, 29, 35, 41, 43, 47, 49, 50.

Fig. 1. Bucharest APA NOVA monitoring points (50 points)

Figure 2 present the municipal distribution system from Timisoara. On the map are marked sampling points for all type of samples.

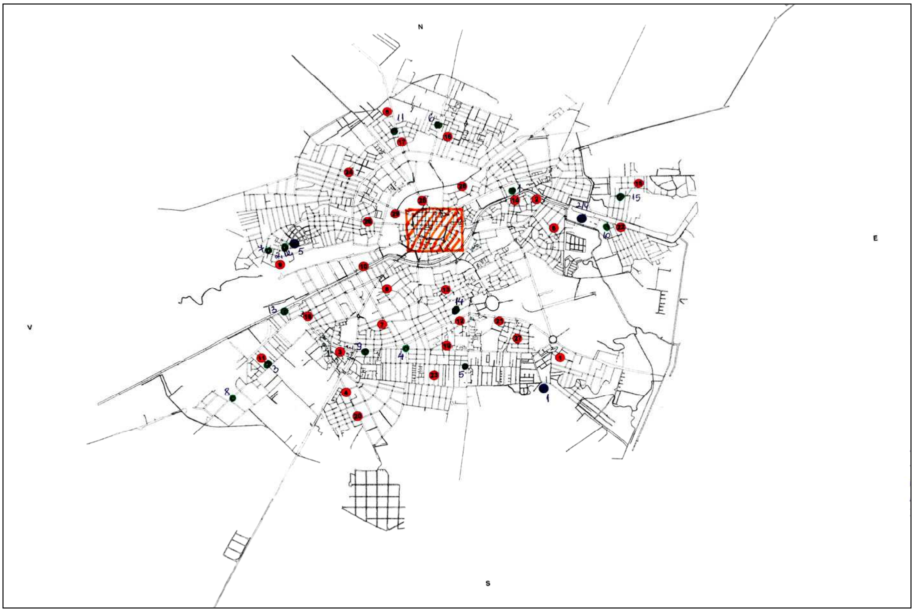
On the Targu Mures map (figure 3) were marked sampling points included in monitoring plan, without random daytime points.

3.3 Materials and methods

3.3.1 Parameters

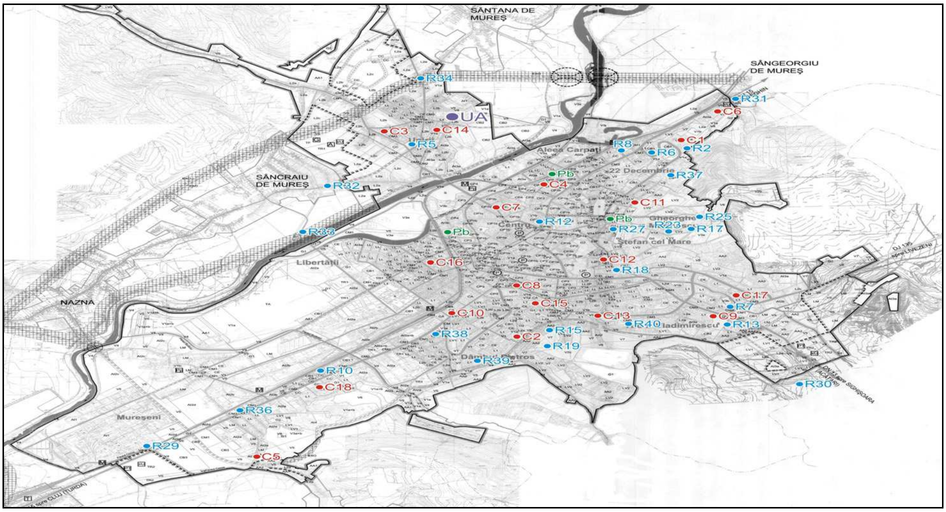
The parameters Al, As, Cd, Cu, Cr, Fe, Mn, Ni, Pb, Se, Sb and Zn were analysed using inductively coupled plasma atomic emission spectroscopy ICP-EOS technique.

In tables 3, 4 are presented the main performance parameters (limit of detection, limit of quantification, accuracy, precision, the uncertainty of measurement) obtained with the equipment and analytical methods used in the study and the maxim admissible value for the metal concentration according to Romanian Legislation (Romanian Law 458, 2002).



Legend: Indigo dots – Water Plants (Bega 1, Urseni 2/4, Ronat 5); Green dots (15 points) - Aquatim monitoring points; Red dots (30 points) - Customer monitoring points (fully flushed and first draw samples); Orange Square (32 points) - Random Daytime samples, historical center of Timisoara.

Fig. 2. Municipal network system in Timisoara City



Legend: Indigo dot – Tg. Mures Water Plant; Blue dots (30 points) – Aquaserv monitoring points; Red dots (18 points) - Customer monitoring points (fully flushed and first draw samples)

Fig. 3. Targu Mures Map

Indicator	Al	As	Cd	Cu	Cr	Fe
Max. Value (μg/L)	200	10	5	100	50	200
LOD (μg/L)	1.00	0.05	0.13	0.60	0.50	0.31
LOQ (μg/L)	3.35	0.17	0.43	2.00	1.50	1.00
Accuracy (μg/L)	0.61	0.15	0.04	0.43	0.77	4.50
Precision (μg/L)	4.29	0.83	0.40	4.18	1.93	6.75
U* (%)	12.24	9.09	9.46	10.37	4.58	3.48
Analytical technique	ICP-EOS	ICP-EOS-FIAS	ICP-EOS	ICP-EOS	ICP-EOS	ICP-EOS

* the value represents the expanded uncertanty for a coverage probability of 95%, the coverage factor is k=2.

Table 3. Performance parameters, maxim admissible value and analytical techniques applied in the study (Al, As, Cd, Cu, Cr and Fe)

Indicator	Mn	Ni	Pb	Se	Sb	Zn
Max. Value (μg/L)	50	20	10	10	5	5,000
LOD (μg/L)	0.10	0.95	0.30	0.10	0.10	0.50
LOQ (μg/L)	0.35	3.20	1.00	0.33	0.33	3.70
Accuracy (μg/L)	0.75	0.04	0.03	0.46	0.20	6.20
Precision (μg/L)	1.80	0.55	0.39	0.95	0.67	3.50
U* (%)	4.31	6.64	9.90	10.06	11.21	4.05
Analytical technique	ICP-EOS	ICP-EOS	ICP-EOS	ICP-EOS-FIAS	ICP-EOS-FIAS	ICP-EOS

* the value represents the expanded uncertanty for a coverage probability of 95%, the coverage factor is k=2.

Table 4. Performance parameters, maxim admissible value and analytical techniques applied in the study (Mn, Ni, Pb, Se, Sb and Zn)

3.3.2 Equipments and materials

- Inductively coupled plasma optical emission spectrometer ICP-EOS type Optima 5300 DV Perkin Elmer with Flow Injection Hydride Generation System FIAS 400;
- Simplicity UV System Millipore for Ultra pure water;
- Bi-Distillate System of drinking water type GFL 2104;
- Electric hot places;
- Fume Hood Laborbau System GmbH;
- Suprapure nitric acid 65%, Merck quality;
- Multi-element Standard Reference Material type Quality Control Standard 21, Perkin Elmer, 100 mg/L As, Cd, Cr, Cu, Fe, Mn, Pb, Sb, Se, Zn;

- Uni-element Certified Reference Materials for ICP, 1000 mg/L, CertiPUR, traceable to SRM from NIST, Merck, one CRM for each element;
- Ultra pure water.

3.3.3 Pre-treatment of the samples

The samples were digested with suprapure nitric acid (5 mL for each sample) and concentrated from 150 mL (sample's volume) to 25 mL (Cvijovic et al., 2010; Dumbrava and Birghila, 2009).

3.3.4 Quality Control of the analytical results

For all the analyzed parameters, the laboratory uses standard method (ISO 11885, 2007), reference materials and certified reference materials. The standard method was verified for all metals and the main performance parameters (limit of detection, limit of quantification, linearity, accuracy, precision, selectivity, the uncertainty of measurement) were established with the existing equipment from the laboratory (Tables 3 and 4).

In the study were prepared recovery samples in the same way as real samples, on three different level of concentration, using Standard Reference Material (Quality Control Standard 21), nitric acid and ultra pure water. The recovery percents were situated in the range 94.5% ÷ 114.5%.

The Standard Reference Material was used also for the calibration curve.

The internal control of results were performed using uni-element CRM for each element.

The samples were analyzed in Pollution Control Department of NR&DI ECOIND, which is accredited by RENAR (Romanian Accreditation Association) and follows the requirements of ISO/IEC 17025:2005 standard. The Department has certification with BVQI (Bureau Veritas Quality International) in accordance with ISO 9001/2008 standard and has periodical participation to internal and external audits.

The Department participates every year at tests for the evaluation of its capability by inter-laboratories comparisons (IMEP Belgium, IAWD Germany, CALITAX Spain, IELAB Spain, AQUACHECK LGS, Quality Infrastructure Denmark) for different groups of pollutants (metallic elements, organic compounds, inorganic compounds) from complex matrixes (surface water, waste water, drinking water, soil, sediment, sludge) and the results were included in the accepted range ($-2 \le$ score $Z \le 2$).

4. Results and discussions

The quality of raw water used by the Companies in the monitoring campaigns indicates some problems.

In winter season, were detected relatively high contents of organic load (COD - 33÷43 mgO_2/L, BOD - 13÷15.5 mgO_2/L) and Kjeldahl nitrogen (3.1÷4.2 mg N/L) in surface water from Bega River. In summer time, were measured high load of iron (3.6÷4.2 mg/L) and Kjeldahl nitrogen.

In addition, the groundwater used by the Urseni Water Plant has high contents of iron (2.9÷3.5 mg/L), manganese (0.35÷0.45 mg/L) and Kjeldahl nitrogen (3.2÷4.5 mg N/L) in both seasons. The raw water used by the Ronat Plant has also high contents of manganese 0.29÷0.83 mg/L) and Kjeldahl nitrogen (3.1÷7.7 mg N/L).

Mures surface water has in summer season high loads of suspended maters, Kjeldahl nitrogen and phosphorus compounds.

In all investigated periods, the quality of drinking water provided by the Operators (APA NOVA Bucharest, AQUATIM and AQUASERV Companies) was situated in the limits imposed by the Romanian Legislation at their responsibility limit (water connections).

The results recorded for metals in customer's tap water were compared with maximum admissible values according to Romanian Legislation (Romanian Law 458, 2002).

The experimental data shows that concentrations of As, Cd, Sb, Se were situated below the detection limit of the method used (ISO 11885, 2007) to all drinking water samples analyzed from the producer – via distribution system- to the customer's tap.

The data for total chromium shows very low contents, under the limit of detection of the method used (ISO 11885, 2007) or situated close to the limit. For these reasons, only concentrations for seven metals: aluminium, copper, iron, manganese, nickel, lead and zinc, are presented in the next section.

All the results for Cu concentrations reported in this study were situated under the EU Drinking Water Directive (2,000 µg/L), but some of them were higher than the Romanian Legislation limit (100 µg/L).

4.1 Statistical data

In order to obtain more informations, the experimental data were statistically procesed using Excel 2003 Microsoft Programme. Thus, for each set of results (ascendent arrangement) were calculated minimum, maximum, mean, median values, standard deviation of the results, percent of non-compliance samples.

In order to compare the results obtained in the study in all three municipalities, the data were organized in accordance with sampling procedure: first draw, fully flushed and random daytime results.

4.1.1 First draw data

In table 5 are presented experimental data obtained from 71 first draw samples collected in June 2009 in Bucharest. The data shows that the limits, were exceed in some samples by several metals: Al, Cu, Fe, Mn, Ni and Pb. A high percent (around 34%) of samples has Fe content above the limit. High percent of non-compliance samples were recorded for Ni (14%) and Cu (10%).

The tables 6 and 7 present first draw data obtained in winter time, respectively in summer in Timisoara. 30 samples were analized and the data show similarly results, such as 14 – 15 samples (around 50%) has metals (Cu, Fe, Ni, Pb) which exceed the limits.

The tables 8 and 9 shows the data obtained in Targu Mures City in winter (35 samples) and summer time (17 samples). In first draw samples were detected high contens of Cu, Fe, Ni (in winter) and Pb (in summer) and the percentage of non-compliance samples was situated in the range 29-43%.

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	3.05	< 0.6	11.1	1.22	< 0.95	< 0.30	1.73
Maxim value	**335**	**621**	**5,676**	**520**	**80.1**	**39.0**	2,194
Median value	42.4	16.0	122	6.73	1.95	< 0.30	250
Mean value	57.3	49.7	271	15.9	9.30	2.81	444
Standard deviation	58.3	112	682	61.1	15.6	6.32	479
Maxim admissible value	200	100	200	50	20	10	5,000
No non-compliance samples / element	3	7	24	1	10	4	0
Total non-compliance samples	34						
Total non-compliance samples (%)	47.89						

Table 5. Bucharest, First Draw Data, June 2009 (µg/L)

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	22.4	4.72	10.7	0.10	< 0.95	< 0.30	0.50
Maxim value	**735**	**397**	**1633**	49.7	**23.2**	**22.4**	1,881
Median value	82.1	24.3	65.5	4.50	< 0.95	1.54	88.2
Mean value	101	80.7	186	7.23	5.21	4.20	321
Standard deviation	118	111	365	9.32	5.03	5.32	449
No non-compliance samples / element	1	9	5	0	1	5	0
Total non-compliance samples	15						
Total non-compliance samples (%)	50.0						

Table 6. Timisoara, First Draw Data, February 2010 (µg/L)

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	17.8	5.43	7.52	1.0	< 0.95	< 0.30	0.50
Maxim value	199	**1029**	**589**	20.1	**32.7**	**36.1**	1,570
Median value	28.4	35.0	60.1	3.92	< 0.95	< 0.30	340
Mean value	40.5	125	106	6.73	4.82	4.51	470
Standard deviation	37.1	210	130	5.31	8.64	9.52	398
No non-compliance samples / element	0	9	4	0	2	3	0
Total non-compliance samples	14						
Total non-compliance samples (%)	46.67						

Table 7. Timisoara, First Draw Data, June 2010 (µg/L)

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	19.2	2.51	<0.31	<0.1	<0.95	<0.30	5.22
Maxim value	78.5	**235**	**732**	24.1	**35.5**	7.2	1,224
Median value	31.6	21.8	62.5	4.14	<0.95	<0.30	245
Mean value	33.8	47.0	154	7.21	5.04	2.36	355
Standard deviation	11.7	59.2	201	5.80	8.74	1.13	339
No non-compliance samples / element	0	4	9	0	3	0	0
Total non-compliance samples	15						
Total non-compliance samples (%)	42.86						

Table 8. Targu Mures, First draw Data, February 2011 (µg/L)

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	21.5	3.11	18.9	1.41	< 0.95	<0.30	8.22
Maxim value	**300**	**244**	**958**	22.0	19.3	**19.5**	365
Median value	38.9	19.5	58.4	3.80	1.35	<0.30	126
Mean value	60.3	49.8	189	5.73	3.32	3.91	169
Standard deviation	65.9	73.3	259	5.40	4.76	4.30	120
No non-compliance samples / element	1	2	4	0	0	1	0
Total non-compliance samples	5						
Total non-compliance samples (%)	29.41						

Table 9. Targu Mures, First Draw Data, June 2011 (µg/L)

4.1.2 Fully flushed data

When the tap was flushed approximately 5 minutes and then the samples were collected, were recorded lower metal concentrations and the percentage (4.2 – 13.3%) of non-compliance samples decreased (tables 10 -13).

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	11.2	<0.6	7.25	<0.1	< 0.95	< 0.30	< 0.5
Maxim value	174	63.2	**1,387**	**96.9**	17.6	5.23	1,453
Median value	36.1	2.52	27.5	2.95	< 0.95	< 0.30	24.8
Mean value	41.8	5.65	63.2	5.67	2.66	0.51	75.8
Standard deviation	22.8	10.7	168	11.6	4.25	0.92	194
No non-compliance samples / element	0	0	3	1	0	0	0
Total non-compliance samples	3						
Total non-compliance samples (%)	4.23						

Table 10. Bucharest, Fully Flushed Data, June 2009 (µg/L)

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	22.7	2.52	15.2	3.90	< 0.95	< 0.30	0.5
Maxim value	**329**	52.8	**294**	12.5	3.03	**11.6**	873
Median value	82.9	6.32	36.1	4.31	< 0.95	< 0.30	3.62
Mean value	90.1	8.80	68.6	6.22	< 0.95	2.21	47.3
Standard deviation	53.7	9.21	68.2	4.63	0.61	3.04	152
No non-compliance samples / element	1	0	3	0	0	2	0
Total non-compliance samples	4						
Total non-compliance samples (%)	13.33						

Table 11. Timisoara, Fully Flushed Data, February 2010 (µg/L)

In table 14 were presented fully flushed results from Targu Mures obtained in summer campaign. The data shows o relatively high percentage of non-compliance samples (around 29%), but the number of analyzed samples was lower than in winter (17 samples, respectively 35 samples).

The metals who exceeded the limits are usually iron and lead and only occasionally aluminum, copper and manganese (each, one sample).

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	15.1	1.81	7.13	1.32	< 0.95	< 0.30	0.52
Maxim value	63.5	69.5	159	20.8	< 0.95	**12.1**	151
Median value	31.7	5.60	26.1	3.10	< 0.95	< 0.30	15.2
Mean value	34.9	11.2	41.8	4.24	< 0.95	1.61	32.8
Standard deviation	13.5	15.1	45.2	4.02	< 0.95	3.20	38.1
No non-compliance samples / element	0	0	0	0	0	2	0
Total non-compliance samples	2						
Total non-compliance samples (%)	6.67						

Table 12. Timisoara, Fully Flushed data, June 2010 (µg/L)

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	20.4	1.73	<0.31	1.42	<0.95	<0.30	< 0.50
Maxim value	74.3	53.8	**241**	18.6	15.7	4.23	1,124
Median value	32.2	4.74	25.1	4.35	<0.95	<0.30	25.4
Mean value	35.4	11.2	50.1	6.04	2.17	2.10	101
Standard deviation	9.89	12.9	56.7	4.46	3.44	0.41	215
Maxim admissible value	200	100	200	50	20	10	5,000
No non-compliance samples / element	0	0	2	0	0	0	0
Total non-compliance samples	2						
Total non-compliance samples (%)	5.71						

Table 13. Targu Mures, Fully Flushed Data, February 2011 (µg/L)

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	28.2	2.32	32.2	1.23	< 0.95	<0.30	12.3
Maxim value	112	**156**	**429**	15.4	3.72	**12.6**	648
Median value	44.1	4.63	63.1	3.28	< 0.95	<0.30	26,3
Mean value	49.5	24.4	113	5.64	1.58	2.65	96.4
Standard deviation	19.6	40.3	117	6.49	< 0.95	<0.30	13.2
Maxim admissible value	200	100	200	50	20	10	5,000
No non-compliance samples / element	0	1	3	0	0	1	0
Total non-compliance samples	4						
Total non-compliance samples (%)	23.53						

Table 14. Targu Mures, Fully Flushed Data, June 2011 (μg/L)

Fully Flushed procedure was used for the samples collected from monitoring points of the Operators. Therefore, the tables 15 to 19 present the statistical data for these samples.

The data show a low percentage of non-compliance samples for some of the campaigns, such as Bucharest, Timisoara in summer and Targu Mures in winter monitoring programs (4.4 – 13.3%). The concentrations of Fe and Pb were reported in same samples above the limits. Other metals that exceed the limit were aluminum (1 sample, Timisoara, winter), copper (3 samples, Targu Mures, summer) and nickel (1 sample, Targu Mures, summer).

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	27.3	<0.6	<0.31	<0.1	< 0.95	< 0.30	< 0.5
Maxim value	106	23.6	138	18.7	11.1	**31.2**	347
Median value	48.5	1.75	12	5.7	< 0.95	< 0.30	6.25
Mean value	51.2	4.11	29.1	5.88	1.34	1.81	48.2
Standard deviation	18.8	5.1	36.3	4.5	2.10	6.46	93.2
Maxim admissible value	200	100	200	50	20	10	5,000
No non-compliance samples / element	0	0	0	0	0	1	0
Total non-compliance samples	1						
Total non-compliance samples (%)	4.35						

Table 15. Bucharest, June 2009 (μg/L), 23 samples (monitoring points of Operator)

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	22.7	2.51	15.2	3.93	< 0.95	< 0.30	0.50
Maxim value	**329**	52.8	**294**	12.5	3.00	**11.6**	873
Median value	82.9	6.32	36.1	4.33	< 0.95	< 0.30	3.64
Mean value	90.1	8.84	68.6	6.22	< 0.95	2.21	47.3
Standard deviation	53.7	9.21	68.2	4.63	< 0.95	3.04	152
Maxim admissible value	200	100	200	50	20	10	5,000
No non-compliance samples / element	1	0	3	0	0	1	0
Total non-compliance samples	4						
Total non-compliance samples (%)	26.67						

Table 16. Timisoara, February 2010 (μg/L), 15 samples (monitoring points of Operator)

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	17.8	1.81	19.5	2.04	< 0.95	<0.30	5.80
Maxim value	65.5	80.4	**290**	22.8	2.23	**12,9**	2,681
Median value	26.1	7.41	47.5	5.10	< 0.95	<0.30	83
Mean value	29.3	15.5	81.4	7.31	< 0.95	2.52	338
Standard deviation	12.8	19.7	75.9	6.53	< 0.95	3.72	672
Maxim admissible value	200	100	200	50	20	10	5,000
No non-compliance samples / element	0	0	2	0	0	1	0
Total non-compliance samples	2						
Total non-compliance samples (%)	13.33						

Table 17. Timisoara, June 2010 (μg/L), 15 samples (monitoring points of Operator)

In Targu Mures, in summer time, it was student's holiday. In some monitoring points located in public schools and high schools, were recorded high contents of metals (Cu, Fe and Pb), because drinking water remains a longer time in internal distribution system (table 19).

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	23.2	<0.6	<0.31	<0.1	< 0.95	<0.30	< 0.5
Maxim value	144	18.3	**311**	20.1	18.7	**35.5**	275
Median value	41.3	2.21	60.9	6.20	1.63	<0.30	8.92
Mean value	44.9	3.32	78.1	6.61	3.10	2,54	31.6
Standard deviation	21.2	3.65	56.7	3.31	3.35	4.25	47.7
Maxim admissible value	200	100	200	50	20	10	5,000
No non-compliance samples / element	0	0	2	0	0	1	0
Total non-compliance samples	3						
Total non-compliance samples (%)	5.00						

Table 18. Targu Mures, February 2010 (μg/L), 60 samples (monitoring points of Operator)

Parameter Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	18.1	< 0.6	13.5	1.12	< 0.95	<0.30	3.13
Maxim value	106	**505**	**536**	31.4	**65.6**	**22.5**	818
Median value	24.2	2.10	35.6	8.53	< 0.95	<0.30	18.1
Mean value	27.9	42.3	94.8	7.37	3.15	3.45	104
Standard deviation	15.7	123	138	8.53	11.8	4.50	204
Maxim admissible value	200	100	200	50	20	10	5,000
No non-compliance samples / element	0	3	4	0	1	2	0
Total non-compliance samples	8						
Total non-compliance samples (%)	26.67						

Table 19. Targu Mures, June 2010 (μg/L), 30 samples (monitoring points of Operator)

4.1.3 Random daytime data

In thirty-two samples collected with random daytime procedure from the histhorical center of Timisoara City, 28.13 % are non-compliance samples (highest concentrations than admissible values for Cu, Fe, Mn and Pb) (table 20). These data show that internal distribution systems affect drinking water quality. In order to use a better drinking water, the tap must be washed before the water is collected.

In Random Daytime Program were collected and analyzed 45 samples in Targu Mures (table 21). The experimental data show similarly results with Timisoara campaign, so were detected Cu, Fe, Mn and Pb above the maximum limits in 22% of the samples collected.

The aim of this study was to identify issues related with lead concentrations, but the results show also problems with other metals.

Parameter / Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	13.3	0.91	5.90	1.22	< 0.95	0.30	2.11
Maxim value	67.7	**1064**	**602**	**314**	< 0.95	**16.5**	2,404
Median value	28.0	11.8	38.8	5.03	< 0.95	<0.30	124
Mean value	31.5	57.4	90.3	20.5	< 0.95	2.04	335
Standard deviation	11.9	184	131	56.7	< 0.95	3.22	565
Maxim admissible value	200	100	200	50	20	10	5,000
No non-compliance samples / element	0	3	4	2	0	1	0
Total non-compliance samples	9						
Total non-compliance samples (%)	28.13						

Table 20. Timisoara, June 2010 (μg/L), 32 samples

Parameter / Element	Al	Cu	Fe	Mn	Ni	Pb	Zn
Minim value	15.3	<0.6	3.92	0.71	< 0.95	<0.30	5.92
Maxim value	143	**741**	**1,528**	**1,310**	15.6	**16.4**	1,585
Median value	32.8	6.75	19.3	2.41	< 0.95	<0.30	33.7
Mean value	44.5	58.9	74.1	32.5	3.67	2.38	128
Standard deviation	27.3	147	229	195	5.92	2.15	261
No non-compliance samples / element	0	7	2	1	0	1	0
Total non-compliance samples	10						
Total non-compliance samples (%)	22.22						

Table 21. Targu Mures, June 2010 (μg/L), 45 samples

In table 22 were presented the percentage of samples that exceed the maximum admissible values for all type of samples collected in Bucharest. The results reported for fully flushed procedure indicate a considerable decrease of metal concentrations in tap water.

A relatively high percentages of non-compliance samples were recorded for the samples collected with random daytime procedure (tables 23, 24), such as Cu and Fe.

Municipality	Indicator	The percentage of samples that exceed the maximum admissible values (%)		
		First Draw	Fully Flush	
			Customers	Monitoring points of Operator
Bucharest	Al	4.23	-	-
	Cu	9.86	-	-
	Fe	33.8	4.23	-
	Mn	1.41	1.41	-
	Ni	14.1	-	-
	Pb	5.63	-	1.41

Table 22. Summary of percentage of non-compliance samples for each element - Bucharest

Municipality	Indicator	The percentage of samples that exceed the maximum admissible values (%)						
		First Draw		Fully Flush				Random Daytime
				Customers		Monitoring points of Operator		
		winter	summer	winter	summer	winter	summer	
Timisoara	Al	3.33	-	3.33	-	6.67	-	-
	Cu	30.0	30.0	-	-	-	-	9.38
	Fe	16.7	13.3	10.0	-	20.0	13.3	12.5
	Mn	-	-	-	-	-	-	6.25
	Ni	3.33	6.67	-	-	-	-	-
	Pb	16.7	10.0	6.67	6.67	6.67	-	3.13

Table 23. Summary of percentage of non-compliance samples for each element – Timisoara

Municipality	Indicator	The percentage of samples that exceed the maximum admissible values (%)						
		First Draw		Fully Flush				Random Daytime
				Customers		Monitoring points of Operator		
		winter	summer	winter	summer	winter	summer	
Targu Mures	Al	-	5.88	-	-	-	-	-
	Cu	11.4	11.8	-	5.88	-	10.0	15.56
	Fe	25.7	23.5	5.71	17.65	3.33	13.3	4.44
	Mn	-	-	-	-	-	-	2.22
	Ni	8.57	-	-	-	-	3.33	-
	Pb	-	5.88	-	5.88	1.67	6.67	2.22

Table 24. Summary of percentage of non-compliance samples for each element–Targu Mures

These results shows that the stagnation time of drinking water in internal distribution system even if it is 1, 2 or 3 hours influence the tap water quality in a worse way.

4.2 Correlations between materials and metal concentrations

In tables 25 to 27 are presented the material types inside the customer's building, primary materials in public network and branch pipe in apartments where the metal concentrations were situated above the maximum limits in first draw samples.

Point	Values higher than limits	Household -primary material	Municipal distribution system - primary material	Branch pipe
C1	Cu, Ni	PexAl, Cu	Cast iron	Pb
C2	Fe	PVC, cast iron	Cast iron	Pb
C4	Cu, Ni	PVC, Cu	Cast iron	Pb
C6	Fe	Cast iron, plastic pipe	PEHD	Pb
C7	Fe	Cast iron	Cast iron	Cast iron
C8	Fe	Cast iron	Steel	Pb
C11	Fe	PVC	Steel	Steel
C12	Fe	PVC, cast iron	PEHD	PEHD
C16	Ni	Metallic pipes	Cast iron	Cast iron
C17	Cu	Cu, PE	Cast iron	Cast iron
C18	Fe, Pb	Pb	Cast iron	Pb
C19	Fe, Pb	Pb	Cast iron	Pb
C21	Fe	PVC	Asbestos-cement, Cast iron	Steel
C23	Al, Fe	PVC	Asbestos-cement, Cast iron	Cast iron
C24	Ni	PVC	Cast iron	Cast iron
C25	Al, Fe, Ni	PVC	Asbestos-cement, Cast iron	Cast iron
C27	Ni	Cast iron	Cast iron	PEHD
C31	Cu, Pb	PVC, Cu	Cast iron, Pb	Pb
C34	Fe	Metallic pipes	PEHD	PEHD
C35	Fe	Cu, Pb, galvanized pipe	Cast iron	Cast iron
C37	Ni	PVC	Cast iron	Cast iron
C38	Pb	Pb	PEHD	PEHD
C40	Fe, Ni	Metallic pipes	Cast iron	Cast iron
C42	Fe, Cu	Cu	Cast iron	Cast iron
C43	Fe	Metallic pipes, PExAl	Cast iron	Cast iron
C47	Fe	PVC	Cast iron	Cast iron
C49	Fe, Ni	Plastic	Cast iron	Pb
C50	Fe, Mn, Ni, Zn	Metallic pipes	PEHD	PEHD, Cast iron
C51	Fe	PExAl, Pb	Cast iron	Cast iron
C54	Fe	Cast iron, Pb	Steel	Steel
C58	Al, Fe	PVC, metallic pipes	PEHD	PEHD
C59	Cu, Fe	Cu, cast iron	PEHD	PEHD
C61	Cu	Cu	PEHD	PEHD
C63	Fe	Metallic pipes	PEHD	PEHD

Table 25. Bucharest, source of metals in first draw samples

In Bucharest, a high percent of first draw samples exceeded the iron concentration limit (table 22). Materials used for pipes and fittings in the domestic distribution system in this points are either metallic (Cu, Pb, cast iron) or a combination of plastic and metallic pipes. In addition, the pipes and fittings of APA NOVA Bucharest distribution system are either cast iron, steel or PEHD (high density polyethylene), that's why the effect observed may be a combination of both distribution systems. As was mentioned above (Lehtola et al., 2004b; Sarin et al., 2004), corrosion process in cast iron is the main source for discolouration of drinking water and iron higher content.

Lead contents above the limit are reported in apartments were either the Pb pipes are inside the building or the Pb branch pipes are used to connect municipal distribution network with domestic system.

Source of high levels of copper in first draw samples is only the Cu pipes used for cold water supply in household. The customers have installed central heating in their apartments and hot water circuit was make with copper pipes. In addition, they have replaced the old cold water pipes with copper pipes and can be observed the results, more obvious in Timisoara (Table 26) and Targu Mures (table 27).

Point	Values higher than limits		Household - primary material	Municipal distribution system - primary material	Branch pipe
	Winter	Summer			
C5	Cu	Cu, Ni	Cu	Cast iron	PE
C6	Cu, Pb	Cu, Pb	Cu	Cast iron	Steel
C9	Cu	Cu	Cu	PE	PE
C12	-	Cu	Cu	PE	PE
C13	Cu, Pb	Cu	Cu	Cast iron	Pb
C18	Fe	Fe	Steel	Cast iron, PE	PE
C19	-	Pb	PVC	PE	PE
C20	Pb	Cu, Pb	Cu	Steel	Pb
C21	Cu	-	Steel	PE	PE
C22	Cu, Pb	-	Cu	PVC	Steel
C25	Ni	Ni	Steel	Cast iron	Steel
C27	Cu, Pb	Cu	Steel	PVC	Steel
C29	Fe	-	Steel	PE	PE
C30	Cu	Cu	Cu	Steel	Steel
C31	Al, Fe	Fe	Steel	Cast iron	Steel
C33	Fe	Fe	Steel	Cast iron	Steel
C35	Cu, Fe	Cu, Fe	Cu	Cast iron	Steel

Table 26. Timisoara, source of metals in first draw samples

In Timisoara (table 26) were reported higher values than the limits in first draw samples for Cu, Fe, Ni and Pb.

The sources for lead contents higher than the limit are either branch pipes or PVC pipes. The stabilizers used for PVC production are often composed of salts of metals like lead and cadmium (Kim, 2001), in order to provide properties needed in the final product.

High content of nickel, was observed in same samples (table 25 to 27), which was generated, probably, by cheap taps with poor quality.

As we can see in table 27, source of Fe in first draw samples could we also unprotected steel, used in both distribution systems.

Point	Values higher than limits		Household - primary material	Municipal distribution system - primary material	Branch pipe
	Winter (2 days)*	Summer			
C1	Fe /2	Fe, Pb	PP	PE	PE
C2	Fe /1	-	Cu	Asbestos-cement	Steel
C3	Cu, Ni /1	-	Cu	Steel	Steel
C4	Fe /2	Al, Fe	Cu	Steel	Steel
C7	Cu /2	Cu, Fe	Cu	Steel	Steel
C8	Ni /2	Cu	PE + Steel	PE	PE
C10	Cu /1	-	Cu	Steel	Steel
C12	Fe /2	-	Steel	PE	PE
C16	Fe /2	-	Steel	PE	PE
C18	-	Fe	Steel	PE	PE

*the monitoring programme includes two consecutive days of sampling.

Table 27. Targu Mures, source of metals in first draw samples

5. Conclusions

This research activity demonstrates that materials used in water distribution systems are part of the overall treatment process that affect the water quality which consumers drink at their tap. The interaction between water and the infrastructure used for its supply are fundamental in producing safety drinking water. Subtle reactions between water and different materials used for its transport can affect the finale quality delivered to consumers.

The study, developed in the period June 2009 – June 2011, in a project regarding safety of drinking water distribution systems in some municipalities from Romania, demonstrated that materials used in drinking water domestic installations have a major contribution in the deterioration of water quality supplied by local distribution operators.

In some locations, in first draw samples collected in the morning from kitchen cold taps were detected high quantity of Cu, Fe, Ni and Pb correlated with materials used in internal distribution system (Cu pipes, Pb pipes, PVC pipes, branch pipes, cast iron and unprotected steel pipes, Ni-Cr plated taps). The main causes are the process of water stagnation and the lack of maintenance of the internal distribution materials.

For samples were collected with fully flushed procedure, the quality of drinking water was better, the number of non-compliance samples decreased with 50%.

The customers were advised that, it be not recommended to use the first draw water for cooking and drinking purpose. Recommendations in cases of exceeding the limit values of metals in drinking water were either flushing water for more than five minutes and then use water for household consumption or replacement of pipes and fittings in both, local or domestic distribution systems.

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