Presence and fate of manganese substances in drinking water supply systems

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A wide range of material sources may potentially contribute to contaminant loads in the potable water. The ability of water supply systems to act as emission control barriers for tap water micropollutants, thereby providing environmental benefits in addition to potable water savings, have not been fully explored. This paper investigates the sources, presence and potential fate of a selection of manganese micropollutants in laboratory scaled water supply systems. All of the investigated compounds are listed under the European Council Directive 98/83/EC on the quality of water intended for human consumption (1998) and World Health Organization Guidelines for Drinking-water Quality (2004). Significant water quality changes are identified. Hydro-chemical modelling indicates that the removal of manganese substances during water treatment is likely to be predominant due to the EC Directive manganese concentration requirement, with only minor contributions to the water supply network. Manganese compounds are resistant to biodegradation and the majority of conventional groundwater treatment plants periodically supply water with manganese residual to the potable water distribution system. Water treatment is unlikely to act as a comprehensive barrier for emission of manganese compounds. Hence, it is important to ensure that other manganese source control options (e. g. pipeline materials, manganese substance emission, and groundwater source control) for potable water supply continue to be pursued in order that emissions of manganese compounds from these sources are effectively reduced and / or phased out as required under the demands of the European Council Directive 98/83/EC on the quality of water intended for human consumption.

Key words: manganese, water supply, water quality, pipelines, users' taps

INTRODUCTION

The presence of metal ions plays a central role in the uptake of minerals from water into humans via the formation of useful compounds (Gladwin et al., 2004). Some metals enhance nutrient uptake into the living organisms (Vosyliene et al., 2010), both as a consequence of the mineral compounds of the water and the ability of the organism to mobilize nutrients from different compounds through the action of different catalytic enzymes (Rivett et al., 2008). In addi-

tion to acquiring essential concentrations, metal ion concentrations are also very important at taking up and accumulating nutrients (Rivett et al., 2008). Unfortunately, this ability includes accumulating of heavy metals (Santamaria, 2008) and undesirable compounds, particularly manganese. This can have important consequences for the retention, mobility and availability of undesirable elements in drinking water supply (Bonadonna et al., 2009). A wide range of potential treatment trains are available for water treatment (Mazeikiene et al., 2010) and reuse, but treatment efficiency data for manganese substances is very limited.

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The importance of metals in nutrient migration in water systems is well documented (Verberk et al., 2009). It has been suggested that heavy metals can act as accumulators of organic matters containing some of organic complexes in pipeline sediments (Cerrato et al., 2006). In the previous work, we showed that manganese in organic compounds contained the biggest part of the total concentration in groundwater enriched with organic compounds (Valentukeviciene et al., 2009). In addition, manganese (particularly of organic origin) was shown to accumulate organic iron against the background of low concentrations of organic compounds. Manganese concentrations in many water supply systems have been found to be 2 to 4 higher than in groundwater (Cerrato et al., 2006). Consequently, the consumption of drinking water from the water supply or private groundwater boreholes that have accumulated large quantities of manganese with high total iron contents (Santamaria, 2008) represents an important pathway by which manganese enters the human organisms system.

The chemical behaviour of manganese could be similar to that of iron, which have rather similar physicochemical properties, e. g. valence and ion diameter (Gerke et al., 2008; Echeverria et al., 2009). Iron has often been used for studies on manganese uptake and seems to be dependent on each other's presence in the water intended for human consumption (Valentukeviciene, 2009). Both iron and manganese show the same uptake dynamics and compete for transport along gradients of concentrations in different compartments of water and organisms (Gladwin et al., 2004). However, the dependency between manganese and iron when taken up by human blood cells is not well understood (Santamaria, 2008). The uptake of manganese is strongly related to the uptake of iron, particularly in blood. Manganese influx into cells and its use of oxygen transporters is discussed in great detail in the review by (Santamaria, 2008).

Rivett with co-authors (2008) summarized the dependency between manganese, iron and nitrogen compounds in groundwater and suggested the possible use of iron as an analogue for predicting manganese behaviour. However, manganese has not showed good correlations with iron in studies of biological metal removal, and it has been suggested that there is an alternative pathway for

manganese uptake into cells (Valentukeviciene, 2009). At the cellular level, manganese is accumulated within cells and is the most important ion in creating catalytic potential.

The objective of the present study was to investigate the behaviour of manganese in drinking water supply pipelines (plastic and steel). The important role that pipeline materials play in manganese uptake in drinking water, in particular its role in manganese transfer between sediments and tap water, require a better understanding of the mechanisms involved. An attempt was made to quantify the uptake and distribution of manganese and iron metals in the raw water – treated water – tap water stages and to study the dependencies between manganese, iron and nitrogen compounds in the various transfer steps. Our starting hypothesis was that total iron concentration in water and concentrations of nitrogen compounds could be used as the indicators of manganese concentrations in drinking water.

MATERIAL AND METHODS

Laboratory scaled pilot equipment

The laboratory scaled pilot equipment was located in a laboratory of the Vilnius Gediminas Technical University (Lithuania). The pipeline at the laboratory was metallic or plastic, and the taps were mainly used for sampling and control procedures. Treated water dominated drinking water supply. The inside pipelines layer consisted of PE (plastic pipes), corrosion layer (25-year-old steel pipes), and zinc layer (new galvanized pipes). These three layers are very common for Lithuanian drinking water supply (Valentukevičienė et al., 2011). The dominant pipeline material is cast iron and takes approximately 56.6% of all analyzed drinking water distribution pipeline materials; steel pipelines take 22.5% and plastic pipelines take 20.7%, the rest is unknown because of the lack of information. All materials of selected pipes were chosen with the purpose of specific evaluation of possible manganese interaction and deposition in drinking water distribution systems (Cerrato et al., 2006). A plastic pipeline was used for the control purpose, because manganese cannot be obtained from this material. For further details about water quality see Valentukeviciene (2009). Seven sampling taps

of a diameter of 15 mm were established 1.5 m apart at the end of each pipe. The dominant water flow velocity was 0.3 m/s and water stagnation was 9 h in the night time.

Pipe sections were designed and constructed from three materials (plastic, 25-year-old steel, and new galvanized steel) with an additional pipe section of plastic to serve as control. These systems helped to determine the variation of water quality, sediment growth, and formation of scaling on pipe surface. Also, these sections allowed obtaining similar conditions for a water distribution system. Each system consisted of a 1.5 m long and 0.015 m inside diameter pipeline, which was connected at both ends with a plastic pipeline to form a closed pipe section. Each section operated at two different conditions; at flow-through during 7 days in order to provide a real operation condition of a distribution system, and at recirculation-flow during each night in order to provide a longer retention time and to increase water-pipe wall interaction; the latter flow condition allows a more clear evaluation of variations in water parameters. The adequacy of the system to operate in any configuration was accomplished through a series of valves. Each system had a pump, flow meter, sampling valve and a set of valves to adjust the operation of the system.

Sampling of water and sediments

The samples of water and pipeline sediments were collected from the 7 sampling sections. Four replicate water samples were collected from each section and directly from sampling taps within each of 7 sections. After the removal of junctions and valves, sediment scratches were taken to a depth of 1.5 m using a cylindrical steel tube of 12 mm in diameter softly adjusted to the pipeline inside wall. These sediments sample were diluted with distilled water till 20 ml into separate flasks and stored in hermetically safe conditions at 4 °C until processed in the laboratory.

Water samples of the 7 different sampling taps collected were filtered of any suspended material, identified to sediment level in the laboratory and the fresh water samples were analyzed for determination of concentrations of manganese, total iron, ammonium ions, nitrates and nitrites (see below). The samples were then analyzed at the International Standards (ISO) and / or European Norms (EN) approved conditions for determination of water turbidity and sediment suspended solids. Solutions of sediment samples taken from pipeline inside walls were also used for chemical analyses. The sediment samples (20 ml) were also diluted using the method described by determination instruction manuals for each indicator.

Chemical analyses

The water quality analyses were made and certain technological parameters were determined for the control and evaluation of water quality change processes using international standard methods: manganese concentration, mg/L; total iron concentration, mg/L; ammonium ion concentrations, mg/L; turbidity, NTU; nitrate and nitrite concentrations, mg/L; and other relevant parameters (e. g. pH, Chemical Oxygen Demands). Raw and treated water samples were analyzed in the laboratory of drinking water, a department within the JSC "Vilniaus Vandenys", Vilnius (Lithuania). In case of any significant differences all samples were analyzed once or twice again.

Suspended solids from pipeline sediment samples were analyzed using International Standard methods, including samples preparation procedures and heating till constant weight.

Statistical analyses

The quantitative results are presented as the arithmetic mean of six independent measurements. Significant differences were removed from the estimations of results and the measurements were analyzed once again. The concentrations of above-mentioned substances were measured 11 times in raw water, treated water, in sampling taps and in the water storage reservoirs. The average concentrations at typical points were estimated. The standard statistical estimation error of the arithmetic average was approximately 11%.

The dependencies between concentrations of different compounds and water quality indicators in different pipes were find out after statistical analyses using Mathcad 2001 Professional software.

RESULTS AND DISCUSSION

Water quality characteristics

Typical water quality parameters for the source water (Antaviliai water field, Vilnius, Lithuania) were

as follows: pH 7.33–7.52; conductivity ~514 µS/cm; Fe_{total} 0.3–2.86 mg/L; NH_4^+ 0.19–0.59 mg/L; Mn 0.13–0.19 mg/L; Na ~13 mg/L; Ca 57.8–74.5 mg/L; and dry residual 306 mg/L. Treated water quality intended for drinking purpose followed the limits of the European Council Directive 98/83/EC and relevant NATIONAL Lithuanian Hygiene Norms. Water organic matter content (following Chemical Oxygen Demands, COD_{Cr}) in the pilot scaled pipelines was also high. The mean COD_{Cr} in the water retention time of 125–999 h was in the range of 39–175 mgO_2/L for treated water, and 33–1255 mgO_2/L for raw water, respectively.

Raw and treated water characteristics values were not correlated to the uptake of manganese in either pipeline sediments or supplied water.

Manganese, total iron and nitrogen compound concentrations in PE and steel pipeline compartments

Concentrations of ammonium ions, nitrates and nitrites in treated water were not significantly different from those in raw water, although the values for all three compounds were slightly higher in steel pipelines (Table 1). Manganese concentrations were significantly higher in both plastic and 25-year-old steel pipelines. There were significant triple and 10 times increases in total iron in 25-year-old steel pipelines compared with plastic

pipes and galvanized steel, minimum and maximum values, respectively. Manganese concentrations varied considerably among samples and no significant differences were found among different plastic and old steel pipelines analyzed.

The concentration ratio data (manganese, defined as concentration in a specific pipeline) followed a similar pattern in plastic and 25-year-old steel pipelines, but the enrichment of manganese compounds in galvanized steel pipeline was more evident, particularly in raw water (Table 2).

Additional water quality characteristics: turbidity, suspended solids, COD_{Cr} and water to sediment transfer factors for the samples analyzed, are shown in Table 3. The concentration ratios of suspended solids for pipelines varied considerably between the water sources supplied. In general, raw water had the lowest values and treated water the highest. The water of treated sources had particularly high values of turbidity.

For COD_{Cr}, oxygen for the dichromatic oxidation of organic compounds studied varied between 42–210 mgO_2/L in plastic pipelines and 33–1255 in 25-year-old steel pipelines with raw water (Table 3).

When comparing concentration ratios of manganese, suspended solids and COD_{Cr}, it emerged that galvanized steel pipelines accumulated greater amounts of manganese from raw water than

Table 1. **Minimum and maximum concentrations of manganese, total iron, ammonium ions, nitrites and nitrates in pipelines with treated water**

Pipe material	Manganese concentration, mg/L	Total iron concentration, mg/L	Ammonium ion concentration, mg/L	Nitrite concentration, mg/L	Nitrate concentration, mg/L
PE	0.18–1.35	0.13–0.38	0.04–0.16	0.02–0.03	2.5–9.3
Galvanized steel	0.06–0.79	0.10–0.50	0.05–0.25	0.02–0.08	2.1–9.7
25-year-old steel	0.12–2.10	0.39–4.81	0.05–0.30	0.03–0.08	2.9–6.9

Table 2. **Minimum and maximum concentrations of manganese, total iron, ammonium ions, nitrites and nitrates in pipelines with raw water**

Pipe material	Manganese concentration, mg/L	Total iron concentration, mg/L	Ammonium ion concentration, mg/L	Nitrite concentration, mg/L	Nitrate concentration, mg/L
PE	0.24–2.56	0.46–5.12	0.02–0.35	0.02–0.17	2.7–11.8
Galvanized steel	0.43–5.52	0.71–5.23	0.06–0.26	0.03–0.14	2.7–9.5
25-year-old steel	0.35–2.45	1.33–5.13	0.07–0.62	0.03–0.09	1.5–7.1

Table 3. **Additional water quality characteristics related to manganese in different pipelines**

Water sources	Raw water			Treated water		
Pipe material	PE	Galvanized steel	25-year-old steel	PE	Galvanized steel	25-year-old steel
Turbidity, NTU	1–3	1–3	11–87	5–200	6–118	9–55
Suspended solids, mg/L	b. d. l.	b. d. l.	38	85	39	42
COD$_{Cr}$, mgO$_2$/L	57–210	116–536	33–1255	42–119	73–175	39–175

from treated water. For example, total iron concentrations in galvanized steel pipelines analyzed from the same samples where water samples and sediments were collected from the same taps were about 10 times higher than the total iron concentrations found in treated water sources. The maximum concentrations of ammonium ions in plastic and 25-year-old steel pipelines were about twice higher than in supplied treated and raw water.

Correlations between manganese, water stagnation and total iron concentration in PE and steel pipelines

The manganese concentration and water retention time values were not correlated to the uptake of residual metal in either old or new metallic pipes, but obtained manganese concentrations in plastic pipes (Fig. 1) were correlated to total iron concentration ($R^2 = 0.93$).

There were no correlations between manganese concentration in supplied water and in either water after a long retention time for new steel pipes and in old steel pipes. Manganese concentrations were related to total iron concentrations, and both elements in different pipes were correlated to linear dependency (total iron concentration: 0.14–4.83 mg/L). There was a close positive linear correlation ($R^2 = 0.74$) between manganese concentration in the water samples from all pipes and ammonium ion concentrations (Fig. 2).

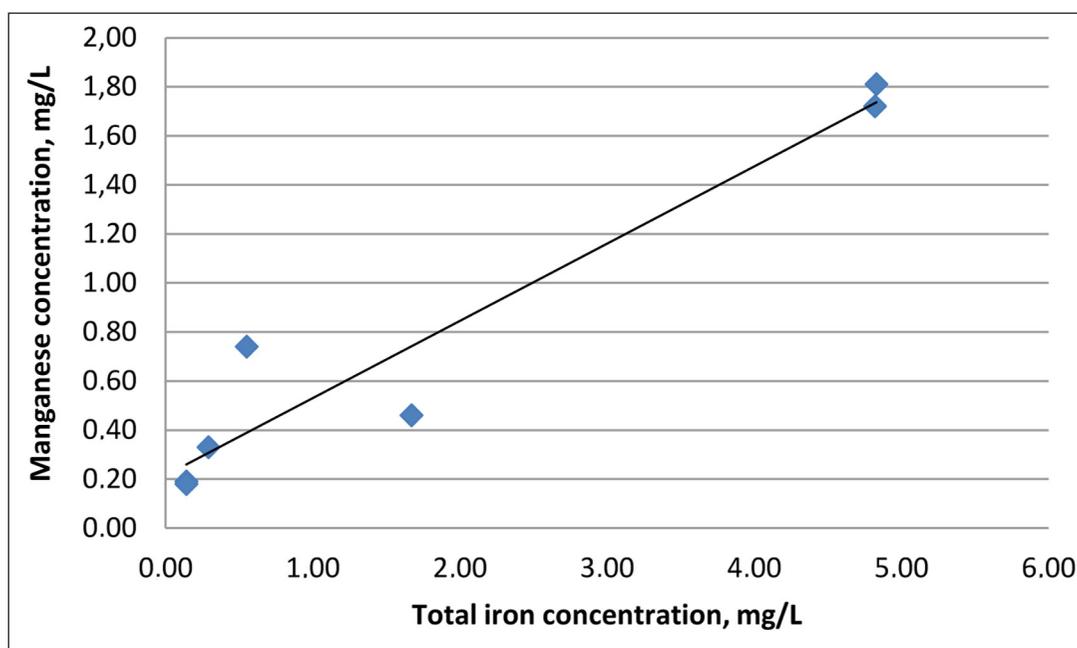The dependencies were absent between manganese and nitrogen compounds (nitrites and

Fig. 1. The correlation of manganese concentration with total iron concentration in a plastic pipe (PE) after long water retention
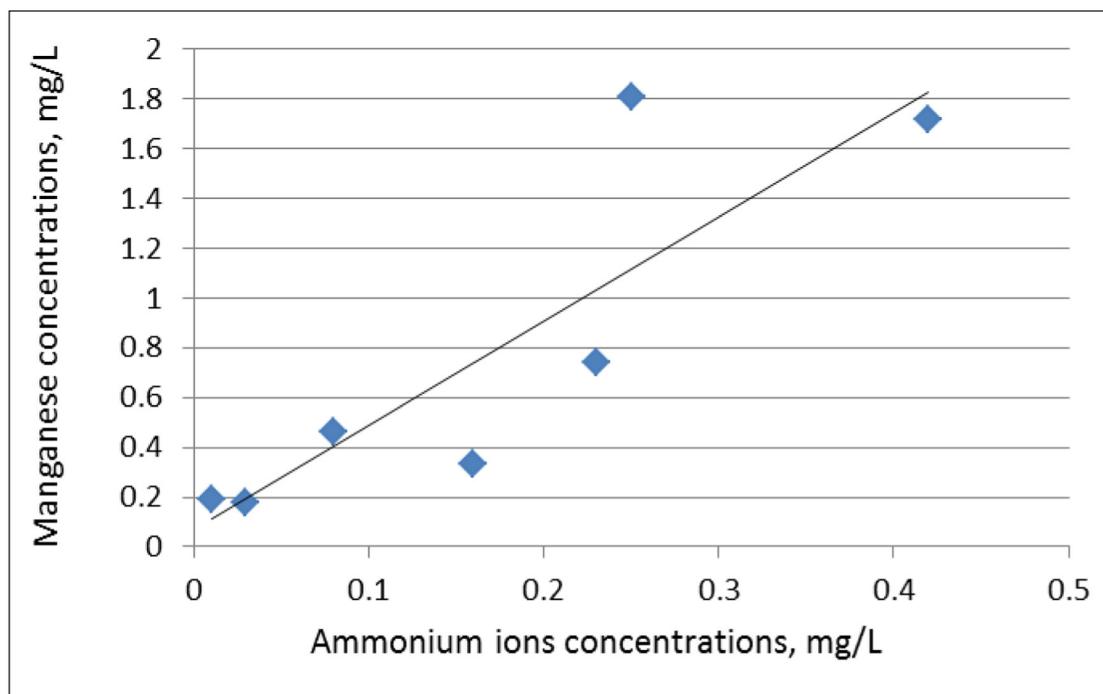
Fig. 2. The dependency of manganese concentration on ammonium ion concentrations

nitrates). The ammonium concentration in old and new steel pipes showed a somewhat different pattern, compared with that in supplied water being closely positively correlated ($R^2 = 0.67–79$) to the water retention time in metallic pipes.

Manganese and total iron concentrations in PE and steel pipelines

Manganese is an essential element for all living organisms, most of which depend on Mn^{2+} uptake, and also eventually on manganese catalyze, for effective metabolism. According to Ljung and Vahter (2007), the current health-based guideline value for manganese in drinking water is based partly on debatable assumptions, where information from previous reports has been used without revising original scientific articles. Manganese deposition in drinking water distribution systems is researched in depth by Ceratto and coauthors, (2006). In the present study manganese composed a greater percentage by weight of the PVC scale than the iron pipe scale; the PVC scale was easily dislodged by flowing water. The high ability of the plastic pipe surfaces to accumulate manganese is well documented. Research by Cerrato and coauthors (2006) has demonstrated that PVC pipe surface accumulates approximately twice higher percentage of manganese weight composition than iron pipes from the same water sources.

Manganese concentrations significantly increased in raw water from galvanized steel pipelines with access to manganese sources both from water sources and pipeline material. Manganese concentrations in galvanized steel pipelines were more than ten times higher than those in raw and treated water sources. The concentration of manganese was nearly the same in old steel pipelines for raw water and treated water.

Total iron concentrations significantly increased in raw and treated water from old steel pipelines with access to iron sources both from water sources and pipeline corrosion material. Iron pipe surface accumulates approximately five times higher percentage of iron weight composition than PVC pipes from the same water sources (Cerrato et al., 2006). Total iron concentrations in galvanized steel pipelines were more than ten times higher in raw water than those in treated water. The concentration of total iron was more than ten times higher in plastic pipelines for raw water than for treated water and thus pipes seemed to accumulate iron from water sources. Similar behaviour was

observed in our previous studies (Valentukeviciene et al., 2009). The different behaviour of manganese and total iron may also derive from their disequilibrium in the content of different pipe materials (Cerrato et al., 2006; Lenthola et al., 2005; McNeill, Edwards 2002; Nawrocki et al., 2010; Sarin et al., 2004).

For the manganese and iron studied, the levels of their concentrations in pipelines after 999 h stagnation were at least ten times higher than those in pipelines after 125 h.

Manganese correlation with nitrogen compounds

Research on uptake mechanisms and attenuation for both metals and nitrogen compounds in anaerobic environment (e. g. water supply pipelines) is fragmented, but some results are reviewed by Rivett and co-authors (2008). They concluded that denitrifying bacteria can obtain energy for metabolism and growth from the oxidation of reduced iron and manganese, metabolic requirements for nitrogen can be met by available NH_4^+ or organic nitrogen, or from the direct assimilation of nitrate. Bacteria also require micro-nutrients (such as manganese and iron). A conceptual model of denitrification occurrence shows the crossroad point when nitrate concentration significantly decreases, manganese concentration slightly increases, thereafter iron concentration reaches the maximum (Berry et al., 2006; Langmark et al., 2005). Our findings indicate that any dependencies of the above-mentioned characters cannot be estimated for plastic or steel pipelines. The preference of manganese or iron metal and nitrogen compounds depends on the nutritional status of the organism, which can explain the differences reported between field experiments (Valentukeviciene et al., 2009) and laboratory experiments with a long water stagnation time.

In the second part of the study we investigated the accumulation of manganese at various water stagnation times and attempted to estimate the dependencies between concentrations of manganese and water turbidity. A significant correlation between manganese concentration and water turbidity may indicate secondary pollution of drinking water with accumulated and removed by water flow manganese (Ramirez et al., 2009). Indeed, Cerrato and co-authors (2006) came to a similar conclusion, suggesting that manganese concentra-

tion in pipelines seemed to be controlled by first flush phenomena regardless of the metal origin (integrated in biofilm or mineral sediment part).

Stable manganese accumulation was not correlated with nitrogen compound concentrations, suggesting that accumulating mechanisms of this metal into pipelines may differ from conventional biological processes.

The results provide some new insights into the use of different pipe materials for drinking water supply purposes or selection of water sources. The transfer of accumulated manganese from pipelines to the users' taps raised some particular solutions about possible prevention mechanisms.

CONCLUSIONS

The concentrations of two metals, manganese and iron, and nitrogen compounds were determined in pipelines of various materials: plastic (PE), galvanized steel, and old steel with a well developed corrosion layer. Water was found to be highly enriched with manganese and iron after a long stagnation time.

Excessive total iron concentration and long retention time in different pipelines could be used as the indicators of increased manganese concentrations in tap water. Increased manganese concentrations seemed to be of integrated physical-chemical-biological origin, and prevention procedures need to be integrated into effective water disinfection, selection of appropriate water sources and pipeline material. This research demonstrates that reactions between water and pipeline material used for its supply can negatively affect the quality of drinking water in users' taps.

The mechanism of manganese accumulation in pipelines could be similar to that of iron, whereas the nitrogen compound accumulation mechanism is different.

Hence, it is important to ensure that other manganese source control options (e. g. pipeline materials, manganese substance emission, and groundwater source control) for potable water supply continue to be pursued in order that manganese compound emissions from these sources are effectively reduced and / or phased out as required under the demands of the European Council Directive 98/83/EC on the quality of water intended for human consumption.

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MANGANO JUNGINIAI IR JŲ SKLAIDA GERIA-MOJO VANDENS TIEKIMO SISTEMOSE

S a n t r a u k a

Geriamąjį vandenį gali teršti daugelis medžiagų. Mikroteršalų sulaikymo galimybės vandentiekio sistemoje nėra visapusiškai ištirtos siekiant papildomai tausoti aplinką ir geriamąjį vandenį. Pasitelkus laboratorinį vandentiekio sistemos modelį buvo tiriami mangano šaltiniai, šio elemento buvimas bei potenciali pernaša ir nustatyti dideli vandens kokybės pokyčiai. Hidrocheminis modeliavimas rodo, kad vandens ruošimo metu mangano junginiai šalinami tik iki lygio, kurį nustato Europos Tarybos direktyvos reikalavimai mangano koncentracijai, ir tik maža jo dalis patenka į vandentiekio tinklą. Mangano junginiai yra atsparūs biologiniam skaidymui, todėl daugelis įprastinio požeminio vandens ruošimo įrenginių periodiškai tiekia vandenį su mangano likučiais į geriamojo vandens paskirstymo sistemą. Vandens ruošimas negali būti pripažintas patikimu mangano junginių sklaidos barjeru. Svarbu užtikrinti, kad tiekiant vandentiekiu geriamąjį vandenį kitos mangano šaltinių kontrolės priemonės (pvz., taikomos vamzdynų medžiagoms, mangano junginių sklaidai ir požeminio vandens kontrolei) būtų nuolat dėmesio centre ir kad mangano junginiai būtų efektyviai mažinami ir / ar šalinami, kaip to reikalauja Europos Tarybos direktyvos.

Raktažodžiai: manganas, vandentiekis, vandens kokybė, vamzdynai, vartotojo čiaupas