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Optimization of NOM Removal during Water Treatment

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Abstract: The aim of this paper is to describe the removal efficiency of individual fractions of natural organic matter (NOM) and the aluminum transformation during treatment of two types of surface water with an increased concentration of NOM of various origins. The coagulation conditions (dose of destabilization reagent and reaction pH value) were optimized for the best NOM and aluminum removal. The results show that the NOM removal efficiency depends on the NOM character, using destabilizing reagents and reaction conditions. The optimized doses of destabilization reagents influence especially the removal of hydrophilic charged (CHA) and very hydrophobic acids (VHA) fractions during treatment of both types of raw water. In contrast to this, the removal of hydrophilic neutral (NEU) fraction is very low ($\varphi_{NEU} = 0.13\text{--}0.22$). The optimal destabilization reagent dosage is characterized by the lowest content of the total reactive aluminum concentration and relatively low concentration of dissolved organic aluminum.

Keywords: Aluminum fractionation, degree of destabilisation, NOM fractionation, particle distribution

INTRODUCTION

Natural organic matter (NOM) such as humic substances, polysaccharides, peptides, amino acids, hydroxyl acids, and others are the common components of surface water (1). Removal of these substances represents one of the basic

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objectives of water treatment because organic matter can be a precursor of organohalogens and other chlorine-derivates (2). NOM can be removed from drinking water by conventional treatment processes as e.g. coagulation or advanced techniques (activated carbon filtration and nanofiltration). The widely used technique is still chemical coagulation (3). The removal of organic matter is best attained by the addition of trivalent Fe or Al salts, which hydrolyze in water and form the hydroxocomplexes. The efficiency of the NOM removal is influenced primarily by the reaction conditions under which the aggregation takes place, the type and dosage of the destabilization reagent and the reaction pH value (4). The reaction pH at which the organic matter is most efficiently removed is determined by the predominant type of organic matter. Since the NOM are a mixture of organic compounds, the efficiency of water treatment is dependent on an effective removal of the main group of NOM. Chemical coagulation preferentially removes hydrophobic NOM fractions over hydrophilic NOM fractions as well as high molecular organic matters are removed more effectively than low molecular organic molecules, that are only partially removed in this process (3).

For a better understanding of which types of organic compounds occur in water before, during, and after the treatment processes, a number of characterization techniques have been developed worldwide. One of them is a fractionation technique with the help of various kinds of resins (5–7). NOM fractionation provides a lot of essential information regarding the structural characteristic of NOM and classifies dissolved organic matter based on their polarity and acidobasic properties (8). The resin adsorption method is used for the isolation of four NOM fractions: very hydrophobic acids (VHA) – humic substances, mainly humic acids; slightly hydrophobic acids (SHA) – humic substances, mainly fulvic acids; hydrophilic charged (CHA) – non-humic base (proteinaceous materials) and acid (an organic compound of the hydroxyl acid group) substances; hydrophilic neutral (NEU) – uncharged matters, mainly polysaccharides (6, 7).

Understanding the distribution and transformation of the aluminum species in the raw water as well as during water treatment processes plays a significant role in the controlling of the drinking water quality. It is well established that the chemical speciation of aluminum is dependent on a variety of chemical and physical parameters, such as pH, temperature, colloidal material, organic and inorganic ligands (9).

The aim of this contribution deals with the NOM and Al fractions removal in various types of raw water by chemical coagulation and compares the efficiency of these fractions removal.

MATERIALS AND METHODS

The studied raw water was taken from the Flaje (northern part of the Czech Republic) and the Vrchlice (middle part of the Czech Republic) drinking

water reservoirs. Both sources contain an increased concentration of NOM of various origin.

The jar tests were applied for optimization of the reaction conditions (dosage of destabilization reagent, reaction pH value) of particle aggregation during treatment of both types of surface water. The aluminum sulphate was used as a destabilization reagent. In order to increase alkalinity, the lime water was used as an alkalization reagent. A variable speed eight-paddle stirrer LMK 8-04 (EJP company, Czech Republic), and 2-liter jars were used for jar testing. The destabilizing reagent was added before a rapid agitation ($t = 1$ minute, $G = 400 \text{ s}^{-1}$). This was followed by a slow agitation ($t = 15$ minutes, $G = 100 \text{ s}^{-1}$) and a settling ($t = 60$ minutes) (4, 10).

DOC (Dissolved Organic Carbon) and COD_{Mn} (Chemical Oxygen Demand) values were used as the principal technological parameters characterizing NOM. UV absorbance at 254 nm (UV_{254}) was measured to provide information about the content of humic material (8). Specific UV absorbance (SUVA) was calculated as a ratio of the UV_{254} to DOC. This rate provides an estimate of abundance of UV absorbing species, and can also be used for comparison of the aromaticity of various NOM samples. Waters with elevated SUVA values exhibit higher hydrophobic/hydrophilic ratios than waters with low SUVA value, this results in greater DOC removal due to chemical coagulation. High SUVA waters (>4) are generally enriched in hydrophobic NOM, such as humic substances (8).

The NOM and aluminum fractionations were used for the diversification of the NOM and aluminum types in raw and treated water. The NOM samples were fractionated by standard resin adsorption technique into VHA, SHA, CHA and NEU components (6, 7), see Figure 1. These fractions were isolated using Supelite DAX-8, Amberlite XAD-4 and Amberlite IRA-958 resins. An applied fractionation technique was carried out according to the method of Chow (7).

$$\begin{aligned} \text{DOC}_{\text{VHA}} &= \text{DOC}_1 - \text{DOC}_2; & \text{DOC}_{\text{SHA}} &= \text{DOC}_2 - \text{DOC}_3; \\ \text{DOC}_{\text{CHA}} &= \text{DOC}_3 - \text{DOC}_4; & \text{DOC}_{\text{NEU}} &= \text{DOC}_4 \end{aligned} \quad (1-4)$$

The method of aluminum fractionation is based on the cation exchange column procedure which is used for separating inorganic from organically bound Al, and the acid digestion method to solubilize Al particulates. The principle of the cation exchange method consists in the existence of inorganic Al in cationic form under the acidic conditions and its retention within the exchange resin, whereas organically bound Al is non-ionic or anionically charged and passes through the column (9, 11).

According to a modified method described by (9), aluminum was fractionated into nine forms. The fractionation procedure allows directly determine five aluminum fractions (see Fig. 2), additional fractions can be obtained by their differences (for filtering a $0.22 \mu\text{m}$ Millipore membrane filter was always used):

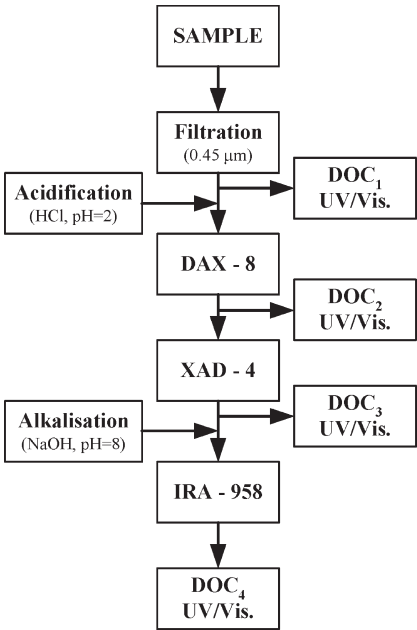


Figure 1. Procedure of NOM fractionation.

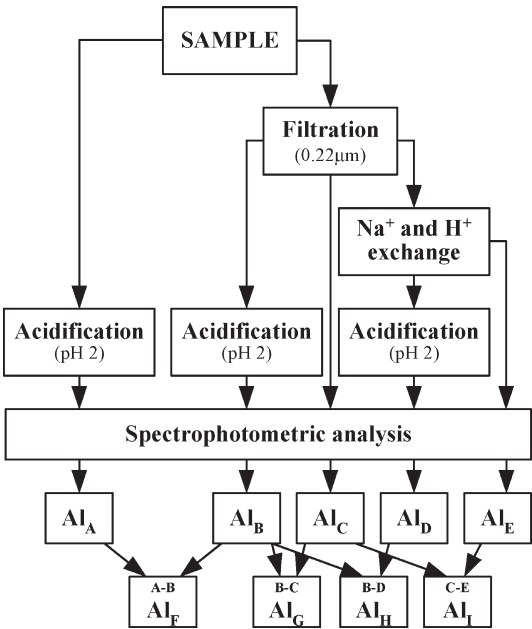


Figure 2. Procedure of aluminum fractionation.

Al_A —total reactive Al (an unfiltered sample was acidified to pH 2 with nitric acid for at least 1 h)

Al_B —total dissolved Al (a sample was filtered and acidified to pH 2 with nitric acid for at least 1 h)

Al_C —dissolved monomeric Al (filtered sample)

Al_D —dissolved organic Al (filtered sample using 0.22 μm Millipore membrane filter, filtrate was passed through a cation exchange column (combined Na^+ and H^+ cation-exchange resin Noromit 001*7) and acidified to approximately pH 2 using nitric acid)

Al_E —dissolved organic monomeric Al (filtered sample, filtrate was passed through a cation exchange column (combined Na^+ and H^+ cation-exchange resin Noromit 001*7)

Al_F —particulate Al (difference between Al_A and Al_B)

Al_G —polymeric-colloidal and strongly bound organic Al (difference between Al_B and Al_C)

Al_H —dissolved inorganic Al (difference between Al_B and Al_D)

Al_I —dissolved inorganic monomeric Al (difference between Al_C and Al_E)

The efficiency of the water treatment process was evaluated by the separation efficiencies of DOC (φ_{DOC}), COD (φ_{COD}) and Al (φ_{Al}). The effect of aggregation processes was evaluated by the degree of aggregation α . The character of the formed suspension was determined by a test of aggregation (sedimentation analysis), enabling the separation of formed aggregates into four basic categories (nonaggregated particles - *NA*, primary particles - *PR*, microparticles - *MI* and macroparticles - *MA*). The analytical processing of the samples and evaluation of the results obtained was described in the previous works (4, 12, 13).

RESULTS AND DISCUSSION

Table 1 presents raw water quality parameters from the Flaje (sampling date 20 Sep 2005) and Vrchlice (sampling date 29 Oct 2005) reservoirs. Total DOC concentrations in the raw water attained $8.19 \text{ mg} \cdot \text{l}^{-1}$ (the Vrchlice reservoir) and $6.61 \text{ mg} \cdot \text{l}^{-1}$ (the Flaje reservoir).

The fractionation results showed that in the raw water from the Vrchlice reservoir, there are dominated NEU and VHA fractions. The raw water from the Flaje reservoir was characterized by predominance of VHA fraction. The SUVA of raw water from the Flaje reservoir was $3.04 \text{ m}^{-1} \cdot \text{mg}^{-1} \cdot \text{l}$, it showed the dominance of hydrophobic humic substances in comparison with raw water from the Vrchlice reservoir ($\text{SUVA} = 1.52 \text{ m}^{-1} \cdot \text{mg}^{-1} \cdot \text{l}$), where dominated mainly hydrophilic non-humic material. The concentration of natural aluminum was found relatively low in the both types of raw water. The dominant aluminum fraction was dissolved organic Al, which formed 66% and 75% of total reactive aluminum in the Flaje and Vrchlice

Table 1. Characteristics of raw water quality from the Flaje and Vrchlice reservoirs

Parameter	Flaje	Vrchlice
pH [–]	6.61	7.72
ACN _{4,5} [mmol.l ^{–1}]	0.20	1.80
COD _{Mn} [mg · l ^{–1}]	6.05	6.18
TOC [mg · l ^{–1}]	6.96	8.83
DOC _{total} [mg · l ^{–1}]	6.68	8.19
DOC _{VHA} [mg · l ^{–1}]	3.61	2.79
DOC _{SHA} [mg · l ^{–1}]	1.28	1.25
DOC _{CHA} [mg · l ^{–1}]	0.06	1.16
DOC _{NEU} [mg · l ^{–1}]	1.73	2.99
UV ₂₅₄ [–]	0.20	0.12
SUVA [m ^{–1} · mg ^{–1} · l]	3.04	1.52
Al [mg · l ^{–1}]	0.095	0.083
Al _A [mg · l ^{–1}]	0.101	0.096
Al _B [mg · l ^{–1}]	0.082	0.081
Al _C [mg · l ^{–1}]	0.058	0.071
Al _D [mg · l ^{–1}]	0.054	0.061
Al _E [mg · l ^{–1}]	0.046	0.057
Al _F [mg · l ^{–1}]	0.019	0.015
Al _G [mg · l ^{–1}]	0.024	0.010
Al _H [mg · l ^{–1}]	0.028	0.020
Al _I [mg · l ^{–1}]	0.012	0.014

reservoirs, respectively. Higher content of dissolved organic Al in the Vrchlice raw water well correspond to higher DOC_{total} and COD_{Mn} concentration in this type of raw water.

The typical jar test results obtained from the optimization of both aluminum sulphate dosage and reaction pH are shown in Fig. 3. With respect to the DOC removal efficiencies the curves show that during treatment of raw water from the Flaje reservoir the optimum reaction pH ranged between pH 5.8 and 6.2 and with increasing pH value residual DOC concentration increased. The highest DOC removal efficiencies during treatment of raw water from the Vrchlice reservoir were obtained in a relatively narrow pH range between pH 5.8 and 6.1.

The treatability of both types of water attainable with different aluminum sulphate dosages are defined in Fig. 4. The highest separation efficiencies ($\varphi_{DOC} = 0.58$, $\varphi_{COD} = 0.74$) during treatment of raw water from the Flaje reservoir were attained using dosages 0.023 mmol · l^{–1} aluminum sulphate and 0.062 mmol · l^{–1} Ca(OH)₂. The maximal removal of organic matter corresponded also to the highest coefficient of aggregation ($\alpha_{Al} = 0.96$).

The results of treatment of raw water from the Vrchlice reservoir showed that the maximal separation efficiencies ($\varphi_{DOC} = 0.41$, $\varphi_{COD} = 0.65$,

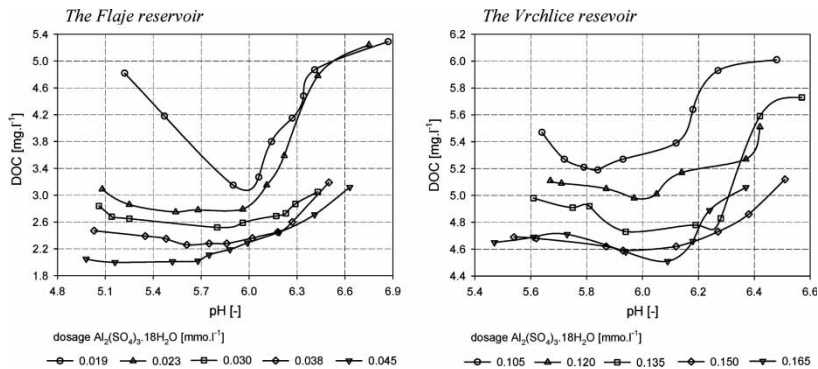


Figure 3. Jar test optimization of aluminium sulphate dosage and reaction pH (the Flaje and Vrchlice reservoirs).

$\alpha_{Al} = 0.90$) were achieved with dosage of $Al_2(SO_4)_3 \cdot 18H_2O$ ($D = 0.135 \text{ mmol} \cdot l^{-1}$), alkalization was unneeded. Both parameters of organic matter, the residual DOC and COD_{Mn} values, decreased with the increasing dosage of aluminum sulphate, though the dosage exceeding $0.135 \text{ mmol} \cdot l^{-1}$ caused only an insignificant increase of the organic matter removal.

Figure 5 presents the results of the test of aggregation. The optimal partial-size distributions of the formed aggregates were observed in the application of previously mentioned optimal doses of reagents. In this case the minimal portions of nonaggregated particles were attained. A predominant portion of the formed aggregates was constituted by the primary- and micro-aggregates ($p = 0.65-0.75$), which are favorable for the one-step separation by filtration (4, 12).

Tables 2 and 3 show the residual concentrations of all DOC fractions during treatment of raw water from the Flaje and Vrchlice reservoirs.

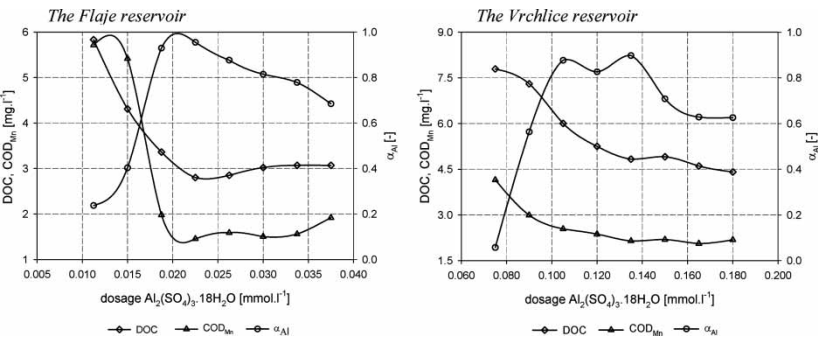


Figure 4. Optimization of dosage $Al_2(SO_4)_3 \cdot 18H_2O$ (the Flaje and Vrchlice reservoirs).

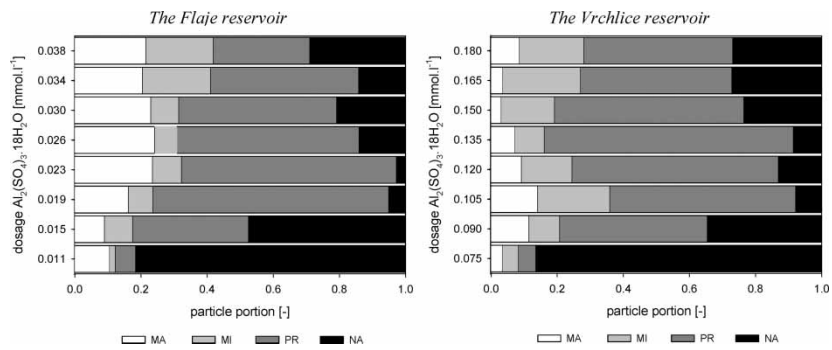


Figure 5. Test of aggregation–size distribution of formed aggregates (the Flaje and Vrchlice reservoirs).

The maximal DOC separation efficiency ($\varphi_{DOC} = 0.58$) during treatment of raw water from the Flaje reservoir was achieved using $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ dosage $D = 0.023 \text{ mmol} \cdot \text{l}^{-1}$. The results of the fractionation show that the residual DOC concentration and distribution of DOC fractions are dependent on the varying destabilization reagent doses.

The highest degree of aggregation ($\alpha_{Al} = 0.90$) during raw water treatment of raw water from the Vrchlice reservoir was achieved for aluminum sulphate dosage $D = 0.135 \text{ mmol} \cdot \text{l}^{-1}$. The maximal efficiencies of DOC removal ($\varphi_{DOC} = 0.46$) were observed in the overdosing aluminum sulphate region (see Table 3).

Figure 6 illustrates changes of the NOM fraction distribution on a value of aluminum sulphate. The highest DOC separation efficiencies of almost all fractions during the raw water treatment of raw water from the Flaje reservoir were observed in the dose of aluminum sulphate ($D = 0.023 \text{ mmol} \cdot \text{l}^{-1}$). The highest DOC removal efficiencies were observed for the VHA ($\varphi_{VHA} = 0.88$) and for CHA fractions ($\varphi_{CHA} = 0.86$). The removal efficiency of SHA attained $\varphi_{SHA} = 0.34$. The NEU fraction was difficult to remove using coagulation process, the maximal removal efficiency was only $\varphi_{NEU} = 0.13$.

The removal of all fractions from the Vrchlice water was less efficient in comparison with the Flaje water and a determination of the optimum dosage with respect to the removal efficiencies of individual NOM fractions was difficult. NEU fraction was the most problematic fraction. The highest removal efficiency of the NEU fraction was determined using of the optimal dose of aluminum sulphate ($0.135 \text{ mmol} \cdot \text{l}^{-1}$), though it was only $\varphi_{NEU} = 0.22$. The removal efficiencies of the other fractions were $\varphi_{VHA} = 0.51$, $\varphi_{SHA} = 0.32$, and $\varphi_{CHA} = 0.78$. Better removal of VHA, SHA, and CHA fractions were observed in an aluminum sulphate overdosing region.

Figure 7 compares the DOC distribution of raw and treated water from the Flaje and Vrchlice reservoirs. The results of DOC fractionation showed that

Table 2. Jar test: concentration of DOC fractions (the Flaje reservoir)

$\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ [mmol · l ⁻¹]	0.011	0.015	0.019	0.023	0.026	0.030	0.034	0.038
DOC _{total} [mg · l ⁻¹]	5.83	4.31	3.36	2.80	2.85	3.02	3.07	3.07
DOC _{VHA} [mg · l ⁻¹]	3.01	1.76	0.53	0.43	0.56	0.59	0.63	0.60
DOC _{VHA} /DOC _{total} [-]	0.517	0.408	0.158	0.153	0.197	0.196	0.204	0.195
DOC _{SHA} [mg · l ⁻¹]	1.12	0.81	1.14	0.85	0.73	0.77	0.73	0.75
DOC _{SHA} /DOC _{total} [-]	0.191	0.189	0.338	0.304	0.255	0.253	0.238	0.243
DOC _{CHA} [mg · l ⁻¹]	0.04	0.04	0.01	0.01	0.02	0.02	0.02	0.04
DOC _{CHA} /DOC _{total} [-]	0.007	0.009	0.003	0.003	0.006	0.007	0.007	0.012
DOC _{NEU} [mg · l ⁻¹]	1.66	1.70	1.69	1.51	1.54	1.64	1.69	1.69
DOC _{NEU} /DOC _{total} [-]	0.285	0.395	0.502	0.540	0.541	0.543	0.550	0.550

Table 3. Jar test: concentration of DOC fractions (the Vrchlice reservoir)

$\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ [mmol \cdot l $^{-1}$]	0.075	0.090	0.105	0.120	0.135	0.150	0.165	0.180
DOC _{total} [mg \cdot l $^{-1}$]	7.79	7.30	6.00	5.25	4.83	4.91	4.60	4.41
DOC _{VHA} [mg \cdot l $^{-1}$]	2.63	2.50	2.06	1.41	1.37	1.40	1.17	1.11
DOC _{VHA} /DOC _{total} [–]	0.338	0.343	0.343	0.268	0.284	0.286	0.254	0.252
DOC _{SHA} [mg \cdot l $^{-1}$]	1.21	1.19	1.07	0.95	0.86	0.90	0.84	0.79
DOC _{SHA} /DOC _{total} [–]	0.155	0.163	0.178	0.181	0.178	0.184	0.182	0.180
DOC _{CHA} [mg \cdot l $^{-1}$]	1.09	0.68	0.42	0.31	0.26	0.21	0.10	0.09
DOC _{CHA} /DOC _{total} [–]	0.140	0.093	0.070	0.059	0.054	0.043	0.022	0.020
DOC _{NEU} [mg \cdot l $^{-1}$]	2.86	2.93	2.45	2.58	2.34	2.39	2.49	2.42
DOC _{NEU} /DOC _{total} [–]	0.367	0.401	0.409	0.491	0.484	0.487	0.541	0.548

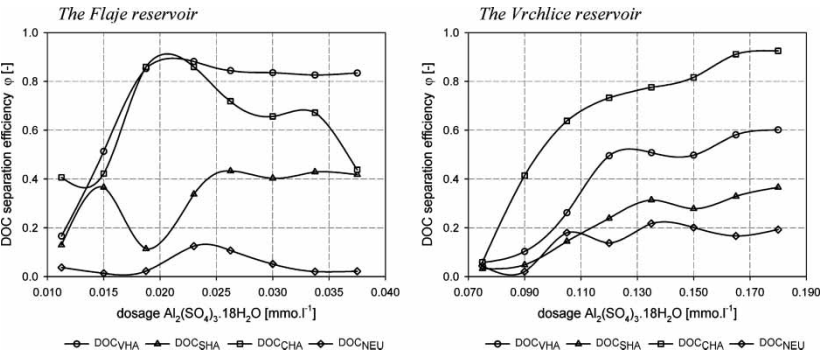


Figure 6. Optimized batch: the efficiency of removal of the individual fractions (the Flaje and Vrchlice reservoirs).

NOM contained in raw water from the Flaje reservoir consisted of 54% of VHA, 26% of NEU, 19% of SHA, and 1% of CHA. The treated water was consisted of 54% of NEU, 30% of SHA, 15% of VHA, and less than 1% of CHA. It was recognized that the VHA fraction predominates in the raw water and NEU fraction predominates in treated water. Based on DOC fractionation of the raw water from the Vrchlice reservoir, it was estimated that there were 34% of VHA, 37% of NEU, 15% of SHA, and 14% of CHA fractions. The NEU fraction also dominated in the treated water from the Vrchlice reservoir, which comprised 49% of DOC_{total}. The portion of other NOM fractions detected in the raw water from the Vrchlice reservoir was the following: 28% of VHA, 18% of SHA and 5% of CHA fractions. From

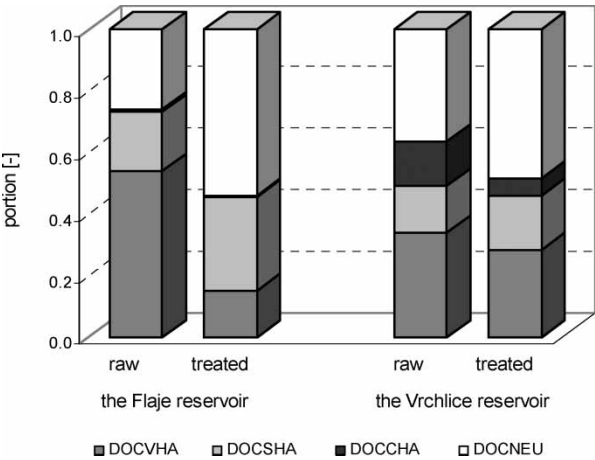


Figure 7. Relative DOC distribution of raw and treated water from the Flaje and Vrchlice reservoirs.

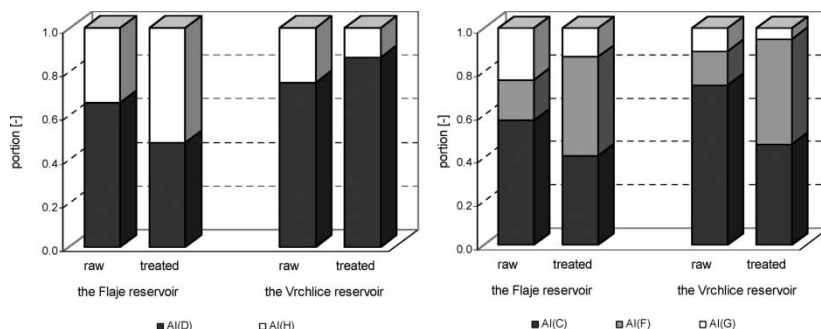


Figure 8. Relative aluminum fraction distribution of raw and optimal treated water from the Flaje and Vrchlice reservoirs.

the results shown in Figs. 6 and 7 it is evident that the chemical coagulation is useful for removal of VHA and CHA fractions but little effective for removal of NEU fraction.

Five aluminum fractions (Al_C , Al_F , Al_G , Al_D , and Al_H) were chosen to demonstrate the different distribution of aluminum in two types of raw and treated water (see Fig. 8). The dominant Al fraction detected in the treated water was the particulate aluminum, which formed 46% and 48% of the total reactive aluminum in the Flaje and Vrchlice reservoirs, respectively.

The fraction of dissolved organic aluminum was found in relatively low concentrations in the treated water from the Flaje reservoir, which correlates to the high removal efficiencies of VHA, CHA and SHA fractions. On the other hand, the main portion of DOC in the raw water from the Vrchlice reservoir was formed by the uneasy removable NEU fraction. This is a reason for evident increase of the concentration of organically bound aluminum in the treated water from the Vrchlice reservoir. The portion of dissolved organic aluminum attained 48% and 87% of the total dissolved aluminum in the Flaje and Vrchlice reservoirs, respectively.

CONCLUSIONS

The results of the residual NOM fractionation showed that the NOM removal efficiency depends on the NOM character and on the operating conditions during water treatment. The conventional coagulation process was relatively suitable for the treatment of surface water with an increased content of organic substances. The NOM fractions in both tested water were slightly different and exhibited the different degrees of treatability. The raw water from the Flaje reservoir was polluted mainly by hydrophobic organic matter of humic character producing colour in water, (73% of total NOM

concentration). The raw water from the Vrchlice reservoir was laden mainly with hydrophilic – not easily removable- organic matter (51% of NOM).

The optimized doses of reagents and pH reaction value especially influenced removal of charged fraction. The hydrophilic charged fraction was the easiest removable, high removal efficiencies ($\varphi_{CHA} = 0.78–0.86$) were observed for both types of treated raw water. Good removal of very hydrophobic acids ($\varphi_{VHA} = 0.51–0.88$) was also observed for both waters. The removal efficiencies of slightly hydrophobic acids ranged from $\varphi_{SHA} = 0.32$ to 0.34 during treatment of both types of raw water.

In contrast to it, it was difficult to remove the hydrophilic neutral fraction (uncharged hydrophilic matter, mainly polysaccharides) using coagulation process, the maximal removal efficiency attained $\varphi_{NEU} = 0.22$ for the raw water from the Vrchlice reservoir and $\varphi_{NEU} = 0.13$ from the Flaje reservoir. Obviously, it is possible to increase removal efficiency of NEU fraction by using further separation step.

The dominant aluminum fraction represented dissolved organic aluminum detected in the raw water from the Flaje and Vrchlice reservoirs. The efficiency of dissolved organic Al removing was especially dependent on VHA, SHA, and NEU fractions. The dominant Al fraction detected in the treated water was the particulate aluminum, which formed 46% and 48% of the total reactive aluminum in the Flaje and Vrchlice reservoirs, respectively.

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