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Removal of precursors of chlorinated organic compounds in selected water treatment processes

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ABSTRACT

This paper discusses the impact of coagulation, ozonation and adsorption on the activated carbon, the content of precursors of chlorinated organic compounds. Coagulation guaranteed the effective elimination of non-biodegradable organic substances and essentially did not impact the biodegradable dissolvent organic carbon (DOC) concentrations. The coagulation process brought about a decrease in concentration for all analysed fractions of organic substances. The removal efficiency of organic substances was higher for mixed water than surface water (SW). Conversely ozonation lowered the biological stability of water by increasing the biodegradable DOC concentration, while only marginally causing the mineralisation of refractive substances. The non-biodegradable organic substances' mineralisation level for the water samples was between 0.00–1.35 $\mathrm{g\,C/m^3}$ (for SW) and 0.02–0.46 $\mathrm{g\,C/m^3}$ (for infiltration water). In general, a higher mineralisation level was registered for SW. Adsorption, despite its substantial removal effectiveness, did not cause the total elimination of high-molecular weight organic substances. In general, total and dissolved organic carbon was more effectively eliminated from SW, probably due to higher pre-adsorption concentration levels in SW. All analysed processes decrease the potential to create chlorinated organic substances through a disinfection process.

Keywords: Organic substances; Water treatment; Coagulation; Ozonation; Adsorption

1. Introduction

Chloride is widely used as a disinfectant in European water treatment plants (WTP) [1], which due to the risk of chlorinated organic substances (TOX) formation constitutes a severe threat to human health [2,3]. Formation of chlorinated disinfection by-products (DBP) only occurs when organic substances have been insufficiently eliminated during water treatment processes proceeding disinfection. The amount and type of DBP are dependent on the type and characteristics of natural organic matter present in treated water. The parameter which determines the potential for the formation of chlorinated organic substances is

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the particle mass of substances found in the water [1,4–6]. The research [7] showed that DBP formation is determined by dissolvent organic carbon (DOC) concentration and the value of UV_{254} is determined as a measure of refractive organic substances [1]. However, compounds with low molecular weight (LMW) have not proved to be susceptible to forming chlorinated organic substances [7]. There is also no unequivocal information as to the susceptibility of hydrophilic or hydrophobic substances towards the formation of DBP. Some authors [1] have concluded that hydrophobic substances are the precursors of chlorinated organic compounds. However, others [8,9] claim that a more significant increase in potential for TOX formation occurs in the presence of hydrophilic substances.

The need to remove precursors of organic substances from water makes it essential to expand the classical water treatment train (coagulation + sedimentation + filtration + disinfection) to include processes delivering more efficient elimination of organic substances. Unfortunately, the widely used coagulation process only guarantees partial elimination of hydrophobic substances [1,10] and a negligible removal of compounds with LMW [11]. Therefore, more and more efficient processes of natural organic matter removal are introduced into the water treatment train, such as adsorption with granular activated carbon (GAC), chemical oxidation or membrane separation [12].

During adsorption, particles with medium molecular weight are effectively removed [13–17]. However, LMW substances, which determine BDOC and assimilable organic carbon (AOC) concentration, are largely unaffected by adsorption. A significant decrease of these biodegradable fractions have only been proven to take place during bio-degradation induced by micro-organisms present in GAC deposits [13,18].

A multitude of research experiments have confirmed the usefulness of ozonation for the removal of precursors of chlorinated organic substances [19–22]. The removal of these substances usually occurs through the transformation of substances with high molecular weight to such with smaller mass, ergo causing the increase in BDOC and AOC concentration. Therefore, the use of the ozonation process proceeding GAC filtration allows for effective biodegradation of these forming fractions, which are easily assimilated by micro-organisms [23].

Ion exchange processes [24] and membrane separation processes (especially ultrafiltration and nanofiltration) [8,25] are less widely used for the removal of TOX precursors. The effectiveness of these processes is, however, comparable to results obtained from the ozonation/GAC filtration sequence.

An analysis of the amount and type of formed TOX indicates that the potential for formation of chlorinated organic substances is directly proportional to the UV_{254} value [7,23,25] and to UV_{254} changes during water treatment processes.

2. Research methodology

Research was conducted at three WTP: for surface water (SW), infiltration water (IW) and for mixed SW micro-sieved with groundwater after aeration (MW). Estimating the removal efficiency of the precursors of chlorinated organic substances was only conducted for selected processes of the water treatment. Therefore, the effectiveness of removal of TOX precursors during processes was specified through technical processes: volume and contact coagulation, indirect oxidation with ozone and adsorption with the use of GAC, which on the basis of available scientific literature [26,27] should guarantee an effective elimination of these substances.

The process of contact coagulation was utilised in the mixed WTP through four accelerators, each with 2,600 m^3 volume. Iron sulphate was applied as a coagulant with doses dependent on the quality of the treating water, which was kept within the range of 13.5–20.3 g Fe/m^3, during conducted research. Water was stored in accelerators for periods ranging 7.8–16.5 h, depending on the temporary output of the water treatment facility.

Volume coagulation was carried out in the SW treatment plant via a labyrinth of rapid mixing chambers (RMC) and slow mixing chambers (SMC), the latter of which are combined with three levels of horizontal-flow sedimentation tanks. Retention times for treating waters in separate installations did not exceed 124–213 s (RMC), 60.2–103.2 min (SMC) and 9.0–15.4 h in the sedimentation tank. Pre-hydrolated polyaluminium chloride is used with doses set at 1.16–4.12 g Al/m^3.

Indirect ozonation is conducted in surface and infiltration WTP. Both facilities utilised this process in ozonation chambers with water retention times verified at 48–82 min for SW and 7–11 min for IW Ozone doses were within the range of 0.51–4.60 g O_3/m^3 (SW) and 0.51–1.00 g O_3/m^3 (IW).

Another process with a confirmed efficiency at removing precursors of chlorinated organic substances was adsorption with the use of GAC. The surface water underwent filtration with the use of WG Gryfskand activated carbon and a contact time with activated carbon estimated at 27.1–46.5 min, whereas IW was in contact with NORIT ROW 0.8 SUPRA active carbon for 27.6–41.3 min.

Research samples were taken before and after each of the analysed water treatment processes with the duration of water flow through separate installations left unchanged. Retrieved water samples were marked with respect to their content in total and dissolved organic carbon (TOC and DOC) as well as biodegradable dissolved organic carbon (BDOC). In order to measure the concentration of aromatic organic substances, the ultraviolet absorbance value (UV_{254}) was determined. Subsequently, the concentration of non-biodegradable dissolved organic substances (NBDOC), used to measure refractive substances, was estimated as the difference in DOC and BDOC concentrations. Additionally, the pH and temperature was measured in the retrieved water samples.

It was accepted that the removal efficiency of precursors of chlorinated organic substances be quantified by the degree, to which NBDOC content and UV_{254} adsorption was lowered through various processes.

TOC and DOC content analysis were conducted in accordance with existing norms with the use of a TOC analyser, whereas BDOC concentration levels were determined via the Van der Kooij method [28] by calculating the content difference in dissolved organic carbon before and after incubating the water with micro-organisms characteristic for the given environment. The other analysed water quality parameters were determined in accordance with current norms.

3. Discussion and research results

3.1. Coagulation

Waters subjected to coagulation characteristically contain a large amount of organic substances (Table 1), for which variability was much higher in SW rather than in mixed one. Both types of water were dominated by DOC, for which TOC fraction was determined to be at 54.2–96.0% for SW and 56.8–99.2% for mixed water. The non-biodegradable fraction, which easily reacts with chlorinated oxidisers, was dominant amongst dissolved organic substances. In both types of raw water, the NBDOC concentration was directly proportional to the ultraviolet absorption value, as depicted by the following equation: $UV_{254} = 2.369*NBDOC + 2.295$ (for SW) and $UV_{254} = 1.622*NBDOC + 11.74$ (for MW). This equation is based on values of UV and NBDOC concentrations in all raw water probes. This correlation was found statistically significant on confidence level $\alpha = 0.05$. This means that aromatic refractive substances were dominant in these waters.

According to the above data, the researched water was dominated by substances with an average or large molecular weight, and therefore constituting precursors to chlorinated organic substances.

The coagulation process brought about a decrease in concentration of all analysed fractions of organic substances. The removal efficiency of organic substances was higher for mixed water (Fig. 1).

It was confirmed that during coagulation of mixed water TOC, DOC and NBDOC were equally efficiently removed, which means that amongst the removed organic substances, the non-biodegradable fraction was dominant, ergo the precursors of chlorinated organic substances. This also confirms that DOC on average constitutes 90.0% of removed TOC. The NBDOC fraction in the removed DOC (Fig. 2) was very high in mixed water and on average was 89.4%.

As a consequence of diminished concentration of non-biodegradable organic substances, the ultraviolet absorption value decreased by 31.7–72.6%. This dependency was directly proportional: $\Delta UV_{254} = 2.244*\Delta NBDOC + 8.001$. The equation is derived from changes in values of UV and NBDOC concentrations in water during coagulation processes. This correlation was found statistically significant for $\alpha = 0.05$. According to Sangyoup Lee et al. [25] and Chiang, Chang, Liang [6], the significant efficiency in decreasing ultra-

Table 1
Value ranges for water quality parameters before and after coagulation

	SW		MW	
Parameter	Before coagulation	After coagulation	Before coagulation	After coagulation
Temperature, ℃	0.5–21.6	0.7–21.6	7.3–16.4	7.3–19.1
pH	7.1–8.0	6.9–7.8	7.0–7.9	6.5–7.1
TOC, g C/m^3	2.61–11.11	1.80–8.45	4.50–9.71	2.02–5.40
DOC, g C/m^3	2.12–9.56	1.50–7.12	3.27–9.54	1.34–5.01
NBDO, g C/m^{3}C	1.80–8.96	1.38–6.65	2.92–9.01	1.19–4.83
BDOC, g C/m^3	0.195–0.876	0.121–0.510	0.182–1.227	0.109–0.862
UV_{254}, m^{-1}	6.42–32.14	4.46–20.50	14.80–27.56	5.46–12.30

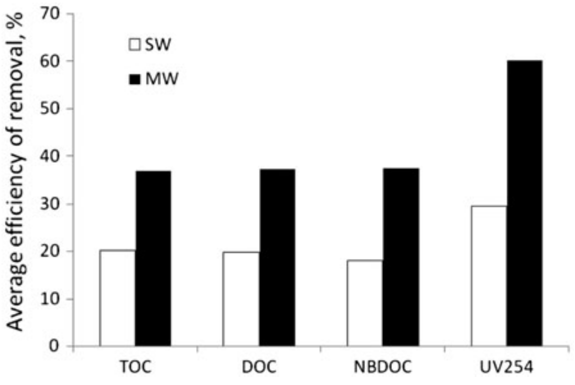

Fig. 1. Average removal efficiency of organic substance fractions.

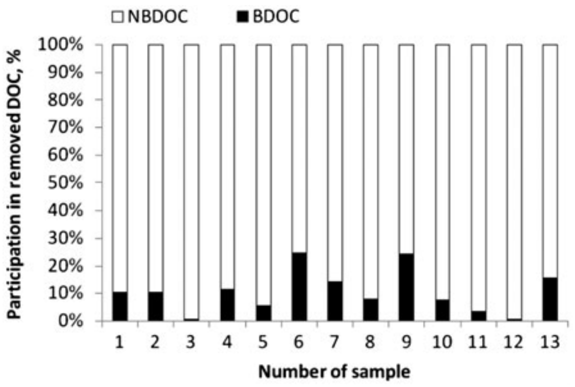

Fig. 2. Organic substance fraction in DOC removed from mixed water.

violet absorbance demonstrates a significant decrease in TOX formation potential.

BDOC removal efficiency, ergo compounds with LMW, was lower than that determined for NBDOC and in 53.8% of the sampled water it did not exceed the analysis error assumed for concentration.

For SW, the NBDOC fraction in removed DOC fell into a very wide range of 2.9–99.7%, dependent on the level of this fraction in raw water DOC (Fig. 3). Consequentially, the BDOC fraction was more pronounced in removed dissolved organic substances. This could be caused by its higher concentration in raw water and the prolonged flocculation process, which was conducive to their adsorption on the surface of post-coagulation sludge. It could also result from different characteristics of BDOC substances in surface and mixed water. As Lin et al. [8] have shown the efficiency of removing organic substances with LMW depends on their structure and hydrophobicity.

It was concluded that for both types of water the level of organic substance content (TOC) removal

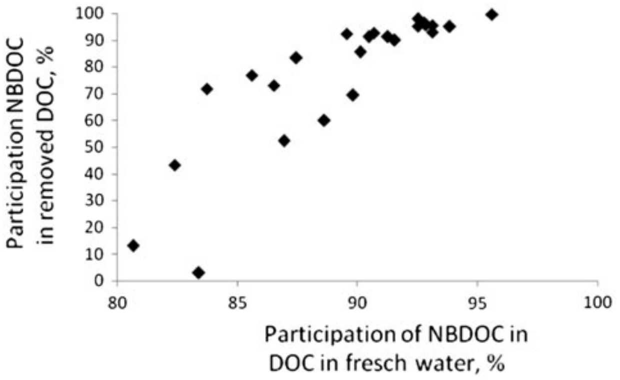

Fig. 3. Relationship between NBDOC fraction in removed DOC and its fraction in raw SW.

improved with a higher removal efficiency of precursors of chlorinated organic substances (Fig. 4) during the coagulation process.

Result analysis showed, conversely to research conducted by Tzoupanos and Zouboulis [27], that organic substance removal efficiency for water was higher when a classical coagulant was used (MW). This can be explained by the significantly higher doses of iron sulphate (g Fe/g C) than of polyaluminium chloride (g Al/g C) as well as by the lower TOC content in raw SW.

Independently of the type of water it was concluded that the decrease in NBDOC concentration was directly proportional to change in UV_{254} absorbance (Fig. 5).

Therefore, independently of the coagulation process methods used, it provided effective elimination of aromatic substances with a greater molecular weight, thus influencing the drop in TOX formation potential

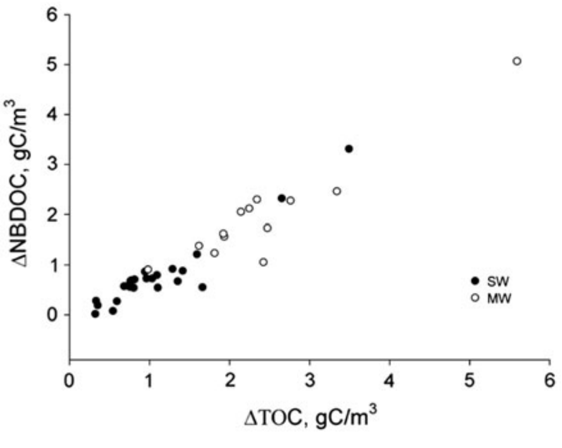

Fig. 4. Relation between decreases in TOC and NBDOC concentration in water as a result of coagulation processes.

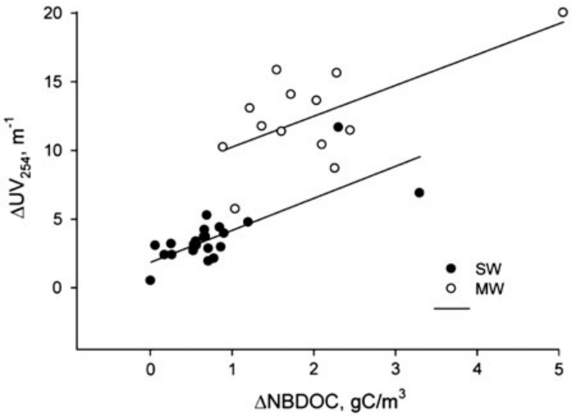

Fig. 5. Relation between changes in NBDOC and UV_{254} concentration levels.

[18]. However, the concentration of organic substances remaining after coagulation (Table 1) suggests insufficient elimination of precursors of chlorinated organic substances through the coagulation process.

3.2. Chemical oxidation

Water subjected to indirect ozonation still contained significant amounts of organic substances (Table 2) with the dissolved fraction being dominant. The NBDOC fraction in dissolved organic substances fell within the ranges of 86.8–95.4% for SW and 79.8–96.5% for IW. In accordance with scientific literature, this should have guaranteed a high removal efficiency of the ozonation process [22,29]. In both types of water a large content of non-biodegradable organic substances coincided with an increased UV_{254} absorbance value.

The ozonation process produced varying results with regards to TOC and DOC removal (Table 3). The efficiency of removing TOC or DOC was determined by the mineralisation of the non-biodegradable fraction of organic substances. This has also been confirmed by other previous research [6,7].

Table 2
Concentration levels of organic substances in water before and after the oxidation process

	SW		IW	
	Before oxidation	After oxidation	Before oxidation	After oxidation
TOC	1.62–6.91	1.25–6.80	1.09–4.66	1.01–4.10
DOC	1.45–5.88	1.15–5.77	0.88–4.55	0.80–4.09
NBDOC	1.27–5.48	0.85–5.02	0.71–4.21	0.50–3.57
BDOC	0.111–0.442	0.278–0.750	0.100–0.375	0.198–0.520
UV_{254}	4.06–15.62	2.82–9.70	3.30–7.98	2.04–5.78

In all surface and IW samples, the decrease in NBDOC concentration as a result of ozonation was greater than the decrease in DOC content (Fig. 6).

This proves the concurrent processes of transforming NBDOC into BDOC as well as the multi-particle mineralisation of organic substances. The share of each of these two processes varied depending on the water; these changes were very significant (Fig. 7).

The NBDOC mineralisation level for given water samples differed within the following ranges of 0.00–1.35 $g\,C/m^3$ (for SW) and 0.02–0.46 $g\,C/m^3$ (for IW). In general, a higher mineralisation level was registered for SW, which could be due to the higher UV_{254} values of this water and the longer time of contact with ozone [6]. As a consequence of lower NBDOC concentration, a UV_{254} absorbance level drop was registered in all water samples. Elimination of TOX precursors during the oxidation process brought about a smaller NBDOC fraction in DOC (Fig. 8).

The confirmed increase in BDOC concentration in both types of water (Table 2), caused by the NBDOC transformation into compounds with lower molecular weight [21,30], effects a decrease in biological stability levels of the water [17], but does not impact a change in potential to form chlorinated organic substances. Ozonation, similarly to coagulation, proved insufficiently effective in the elimination of TOX precursors.

Table 3
Efficiency ranges (%) of organic substance removal

Parameter	SW	IW
TOC	0.0–26.8	1.5–18.1
DOC	0.0–25.5	1.5–14.6

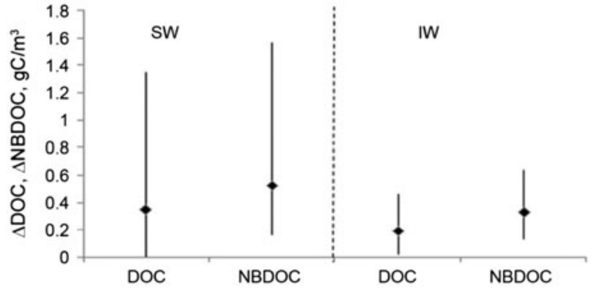

Fig. 6. Ranges and average decrease in DOC and NBDOC content.

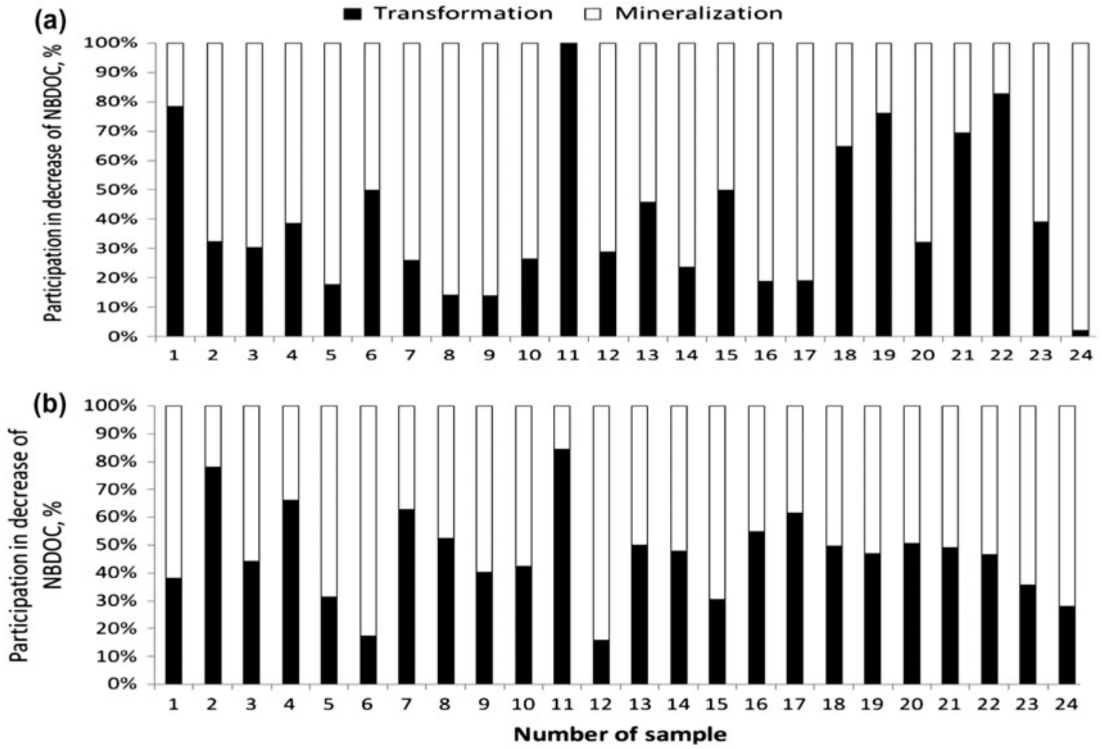

Fig. 7. Share of transformation and mineralisation processes in the decrease of NBDOC levels in (a) SW (b) IW.

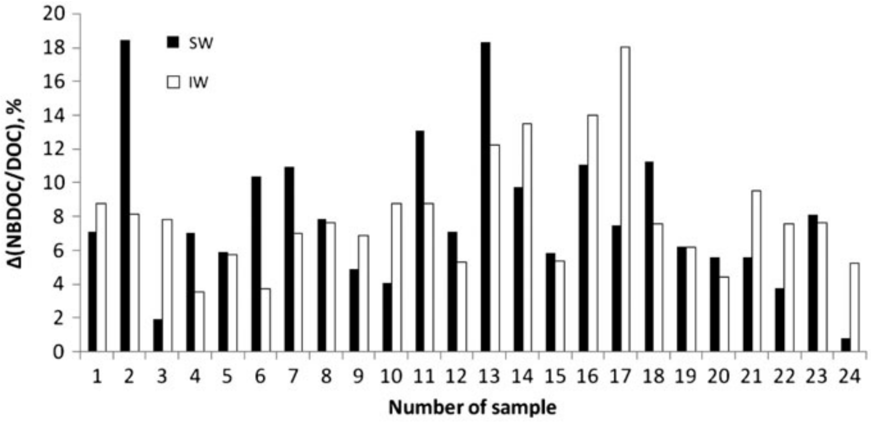

Fig. 8. Decreases participation of NBDOC fractions in DOC.

3.3. Adsorption

The adsorption process was applied in the surface and infiltration WTP. In both types of water, which underwent adsorption, large amounts of BDOC, which were formed as a result of the ozonation process were found to be present (Table 4).

Both types of water were characterised by high fractions of dissolved organic substances in TOC, amounts, respectively, estimated within the range of 43.7–100.0% (for SW) and 46.4–99.8% (for IW). Despite a higher BDOC content than in natural water, NBDOC was dominant amongst dissolved substances (Fig. 9). For 70.8% of water samples, NBDOC levels in DOC were higher in SW than in IW.

TOC removal efficiency in both analysed plants was similar to DOC removal efficiency levels (Table 5).

Table 4
Value ranges for selected water quality parameters before and after adsorption

		SW		IW	
Parameter	Unit	Before adsorption	After adsorption	Before adsorption	After adsorption
Temperature	℃	0.4–21.7	0.5–21.7	8.0–15.3	8.0–15.7
pH		6.8–7.8	6.7–7.6	7.0–7.7	7.0–7.5
TOC	$\mathrm{g\,C/m^3}$	1.23–6.71	0.80–4.32	1.01–4.10	0.76–3.55
DOC	$\mathrm{g\,C/m^3}$	1.15–5.70	0.75–3.82	0.80–4.09	0.40–3.55
NBDOC	$\mathrm{g\,C/m^3}$	0.82–4.93	0.62–3.70	0.50–3.57	0.21–3.28
BDOC	$\mathrm{g\,C/m^3}$	0.230–0.770	0.098–0.345	0.198–0.520	0.064–0.270
UV_{254}	m^{-1}	2.74–9.44	1.16–6.68	2.04–5.78	1.08–4.84

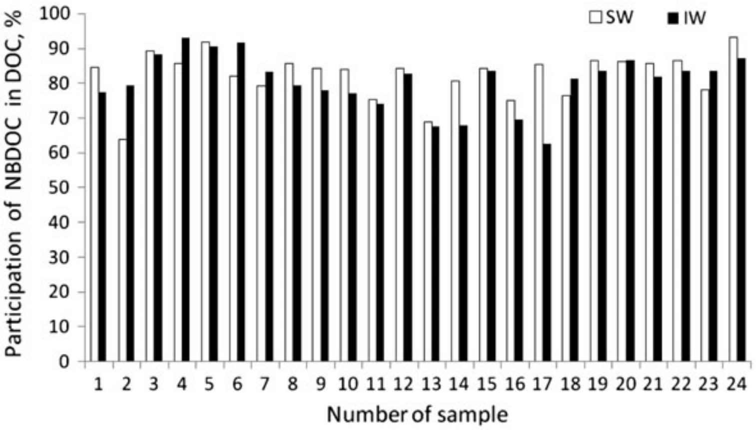

Fig. 9. NBDOC fractions in DOC in water samples before adsorption.

In general, TOC and DOC were more effectively eliminated from SW, probably due to their higher pre-adsorption concentration levels in this water.

High efficiency of BDOC removal from both types of water proves the bio-degradation process initiated by micro-organisms present on an activated carbon surface [13,30], together with the simultaneous adsorption of certain compounds. It has been widely acknowledged that organic substances with LMW are susceptible to adsorption [13,20,31,32]. The participation of precursors of chlorinated organic substances in removed DOC was estimated at 14.7–91.9% (for SW) and 1.8–82.6% (for IW). Lower efficiency of NBDOC removal from IW (as compared to SW) was caused by lower ultraviolet absorbance value in this water and a significantly lower level of organic pollution (TOC). High variation of NBDOC removal efficiency in both types of water brought about a significant post-adsorption increase of the NBDOC fraction in dissolved organic substances (Fig. 10) in the given waters.

Table 5
Efficiency ranges (%) for the removal of organic substances through adsorption

Parameter	SW	IW
TOC	9.2 –54.5	9.3–37.4
DOC	9.0–55.5	7.9–55.6
NBDOC	3.2–54.3	0.2–65.5
BDOC	30.4–84.4	34.6–67.9
UV_{254}	5.6–96.4	16.3–61.9

Furthermore, it was confirmed that the decrease in NBDOC concentration was directly proportional to the decrease in ultraviolet absorbance. This conclusion is based on regression equations: $\Delta UV_{254} = 1.285^{*}$ $\Delta NBDOC + 1.361$ (SW), $\eta UV_{254} = 0.168^{*}$ $\eta NBDOC + 35.2$ (IW). This equations means that NBDOC concentrations changes were determined by refractive organic substances removal. This correlation was found statistically significant from $\alpha = 0.05$.

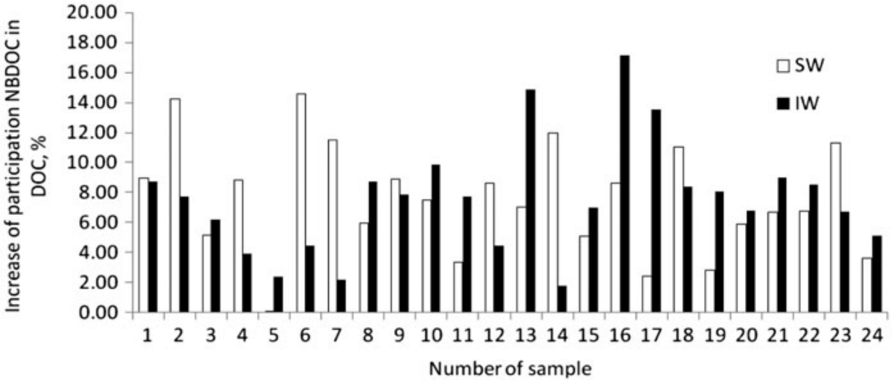

Fig. 10. Increase in NBDOC fractions in DOC.

NBDOC content and the UV_{254} absorbance value for respective waters show that the adsorption process has not guaranteed sufficiently effective removal of precursors of chloride by-products.

Conclusions

Coagulation, irrespective of the implementation method, guaranteed a high removal efficiency of precursors of chlorinated organic compounds as well as, to a lesser extent, the decrease in BDOC content. The efficiency of the coagulation process in NBDOC elimination was, above all, determined by the organic pollution level of water undergoing this process.

Indirect ozonation guaranteed effective elimination of NBDOC, which took the form of a transformation into organic matter with lower molecular weight or via mineralisation. The participation of mineralisation in the removal of non-biodegradable organic substances increased in waters with higher NBDOC content, thus characterised with higher UV_{254} values.

The adsorption process guaranteed the reduction of all fractions of dissolved organic substances, however, due to the presence of biofilm on the surface of activated carbon granules, organic substances with LMW were more efficiently removed than TOX precursors.

The efficiency of removing TOX precursors during elementary water treatment processes was dependent on the type and characteristics of the organic matter present in the water undergoing these processes.

All analysed water treatment processes were found to have decreased the concentration of precursors of chlorinated organic substances. However, none of them guaranteed sufficient removal efficiency of these substances, which would eliminate the potential risk of the formation of disinfectant by-products.

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