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Removal of trihalomethanes from drinking water by nanofiltration membranes

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Abstract

Chlorine reacts with the natural organic matter (NOM) in waters and forms disinfection by-products (DBP). Major of these by-products are trihalomethanes (THM) and haloacetic acids (HAA). They have been known to cause cancer and other toxic effects to human beings. This study determined the removal efficiencies of THM by nanofiltration (NF) techniques with NF200 and DS5 membrane. The rejection of this chlorination by-products was studied at various feed concentration by changing transmembrane pressure. Experimental results indicated that in general increasing operating pressure produces a higher flux but does not have a significant effect on THM rejection. On the other hand, increasing the feed concentration produces a little change in the overall flux and rejection capacity. NF200 membrane removed more THM than DS5 membrane. The higher removal efficiency of dibromochloromethane (DBCM) was attributed to brominating characteristics (higher molecular weight (MW) and molecular size). As a consequence, the results of this study suggest that the NF membrane process is one of the best available technologies for removing THM compounds.
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Keywords: Trihalomethanes (THM); Nanofiltration (NF); Operational conditions; Synthetic water

1. Introduction

Membrane techniques are playing an increasingly important role in the treatment of water sources used for drinking purposes. The greater use of membrane technologies results from more stringent water quality standards, a decrease in adequate water resources, and an emphasis on water reuse [1]. Besides, the relative cost of membrane processes have decreased because of technological advancement, thus prompting their use as increasingly viable water treatment alternatives to conventional treatment methods [2]. Whereas in the past, membrane systems were typically used for desalting purposes only, they are now being used for multiple purposes in the world wide, including desalination, disinfection by-products (DBPs) control, pathogen removal, and removal of inorganic and synthetic organic chemicals (SOCs) [3,4].

Chlorination is the most commonly employed chemical disinfectant in drinking water treatment nowadays. However, several studies reported that chlorination of organic matter in fresh water resulted in formation of DBPs [5,6]. Concerns regarding the potential health effects of DBP prompted several industrialized countries to develop a number of regulations. The disinfectant/disinfection by-product (D/DBP) regulation in United States of America (USA) has set maximum contaminant levels for total THM (TTHM) species (chloroform, bromodichloromethane, dibromochloromethane, and bromoform) and five HAA species (monochloro-, dichloro-, trichloro-, monobromo- and dibromoacetic acid) of 80 and 60 μg/l, respectively [7]. On the other hand, European Union (EU) regulated TTHM limit at 100 μg/l [8]. The WHO guidelines are 200 μg/l for chloroform, 60 μg/l for dichlorobromomethane, 100 μg/l for dibromochloromethane, 90 μg/l for dichloroacetonitrile and

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100 μg/l for dibromoacetonitrile [3]. Moreover, Turkish Government recently regulated 150 μg/l TTHM limit in drinking water to comply with EU regulations [9].

As the limitations of conventional water treatment processes to meet increasingly stringent drinking water regulations become more apparent, membrane processes are gaining support within the water treatment industry as a better means of addressing existing and anticipated regulatory requirements in the world wide. Several researches have focused on nanofiltration (NF) that appears to be promising for controlling DBP in drinking water. In particular, the recently promulgated Turkish standard for THM (150 μg/l) may affect treatment practice of many water treatment plants in countrywide. Thus, Turkish water utilities using surface water as a water source may need to provide additional THM removal processes such as granular activated carbon (GAC) adsorption, or membrane filtration technologies. Membrane processes have been shown to effectively and economically control THM in water [10].

NF now is competitive with other NOM removal technologies of conventional clarification and GAC adsorption because of the following advantages: simplicity of operation, development of higher flux membrane with low fouling potentials, and lower membrane costs [11–13]. Some researchers have reported that membrane costs are comparable to or lower than conventional treatment for small systems of <20,000 m^3/day, or 5 MGD (million gallons per day) [2,14]. On the other hand, there are still a lot of unknowns in the removal mechanisms by NF membranes because of the rejection properties of NF membranes.

Some recent studies have given considerable attention aimed at establishing the removal efficiency of organic solutes by NF from aqueous solutions and hence the mechanism [15]. The removal of organic solutes is influenced by membrane operating variables and membrane sieving effects. Further, certain membrane–solute interactions are believed to be involved. The extent of these influences is largely dependent on the membrane type and permeating solutes, however, the complete principle of organic solutes permeation through NF membranes is unclear. On the other hand, membrane properties such as hydrophobicity, membrane charge, membrane pore size, potential for fouling, resistance to temperature, retention properties and permeability dramatically affect the membrane filtration process. In addition, solute properties of dipole moment and hydrophobic also affect the separation efficiency by adsorbing or interacting with membrane surfaces [4,16,17].

Most organic matters that are responsible for major DBP precursors in Turkish source water are small to medium compounds, with a specific ultraviolet absorbance (SUVA) value varying between 1.5 and 3.0 [18–21], which was consistent with the findings suggested by Amy et al. [14] that the majority of THM formation potential (THMFP) are present in SUVA value less than 3.0. The SUVA parameter represents the ratio UV_{254}/DOC and constitutes an indicator of carbon aromaticity in water.

The objectives of this research were to determine the efficiencies of two membranes (NF200 and DS5) for removing THM compounds. The effects of different pressure levels on filtration mechanisms at various THM feed concentrations were also investigated.

2. Material and methods

The glassware used during analysis was washed with detergent, rinsed with tap water, ultrapure water, and acetone, and dried in an oven at 150 °C for 2 h. On the other hand, methanol (purges and trap grade) and sodium sulfite were purchased from Merck. Ultrapure water was from a Sartorious water purification system (Sartorious). The water samples used in this study were finished water of Terkos Lake water (TLW). Quality parameter of this water is summarized in Table 1. Water samples were placed in the dark in a refrigerator at +4 °C to retard biological activity prior to use.

2.1. Feed water preparation

A stock humic acid solution (sodium salt, Aldrich) was prepared by dissolving 5 g humic acid in 1 l deionized water. Then, this solution was mixed with treated water of Terkos Lake, and filtered by 0.45 μm membrane filter. Further, this resultant solution was chlorinated under standard conditions. Subsequent to reaction period, the residual chlorine was eliminated in the sample using sodium sulfite solution. After that this stock THM solution was diluted with deionized water to get designated THM feed water concentrations of 20, 40, 80, and 200 μg/l, respectively. Table 2 summarizes the properties of THM compounds [22].

2.2. Membrane filtration procedure

The commercial NF200 membrane produced by Dow-FilmTec and DS5 membrane produced by OSMONICS were used in this study. Table 3 summarizes the characteristics of these membranes. A laboratory cross-flow mode filtration apparatus with a flat-sheet membrane cell was used for the filtration tests (Fig. 1). The experimental apparatus were made of stainless steel to avoid undesirable adsorption of the THM compounds. The effective membrane area was 155 cm^2. Fig. 1 is a schematic diagram of the NF module used in this study. Fresh membranes were precompacted at the pressure of 25.0 bar with deionized water. During the experiments, the transmembrane pressure ranged between 5 and 25 bar. Control experiments were also carried out to determine the degree of adsorption of THM compounds onto apparatus. The result shows that adsorption onto the apparatus is negligible. In each experiment, a new membrane was

Table 1
Finished Terkos Lake water (TLW) quality parameters

Variables	Unit	Average values
DOC	mg/l	2.66
UV_{254}	1/cm	0.047
SUVA	l/(mg m)	1.77
Alkalinity	mg $CaCO_3$/l	124
Br^-	μg/l	127
pH	–	7.85
Temperature	°C	19.0
THMFP	μg/l	144

Table 2
Physicochemical properties of THM compounds [22]

Compounds	Molecular weight (g/mol)	Solubility (mg/l)	Log K_{ow}	Henry's law coefficient (Pa m^3/mol)
Chloroform (CFM)	119.4	7500 (25°)	1.97	440
Bromodichloromethane (BDCM)	163.8	3320 (30°)	2.10	160
Dibromochloromethane (DBCM)	208.3	1050 (30°)	2.24	86
Bromoform (BFM)	252.7	3190 (30°)	2.38	54

Table 3
Membrane tested in this study

Parameters	NF200	DS5
Polymer type	Polyamide thin-film	Polysulfone-polyamide
Maximum operating temperature (°C)	45	50
Maximum operating pressure (bar)	41	40
pH range	3–10	2–11
MWCO (Da)	200–300	150–300
Rejection	97% (2000 ppm $MgSO_4$)	80% (2000 ppm Na_2SO_4)

used, rinsed with ultrapure water, and compacted by filtering ultrapure water overnight before starting a filtration test.

2.3. Instrumentation and analytical conditions

DOC measurements were performed with a Shimadzu TOC-5000 analyzer equipped with an auto sampler, according to the combustion-infrared method as described in the Standard Methods 5310 B [23]. The sample is injected into a heated reaction chamber packed with a platinum-oxide catalyst oxidizer to oxidize organic carbon to CO_2 gas. Besides, UV_{254} absorbance measurements were conducted in accordance with Standard Methods 5910 B [23] by a Shimadzu 1601 UV spectrophotometer at a wavelength of 254 nm with a 1 cm quartz cell. Water samples for DOC and UV_{254} were first filtered through a pre-washed 0.45 μm membrane filter to remove turbidity, which can interfere with this measurement, and distilled ultra filtered (DIUF) water was used as the background correction on the spectrophotometer. Further, THM concentrations were determined with liquid–liquid extraction method according to EPA Method 551.1 [24]. The sum of mass concentrations of four trihalomethanes (chloroform, bromodichloromethane, dibromochloromethane, and bromoform) was reported as TTHM in μg/l. Thirty-five millilitres of THM samples were extracted with *n*-pentane, and the extract was then analyzed by gas chromatograph/micro electron capture detector (GC/μECD) (HP 6890) with a fused silica capillary column (J&W Science DB-1, 30 m × 0.32 mm i.d. × 1.0 μm film thicknesses). Duplicate analyses on each sample were performed in accordance with the QA/QC requirement [18,21].

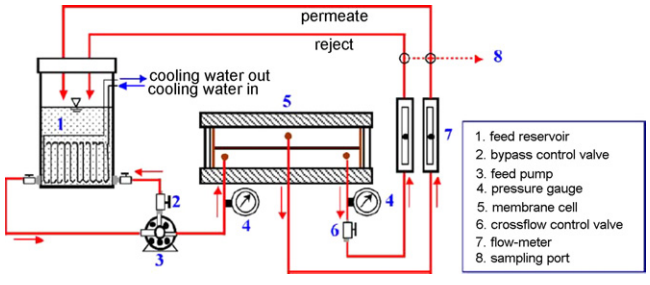

Fig. 1. Schematic diagram of NF membrane unit.

3. Results and discussion

Several people have cited a number of factors affecting removal of organics by NF membranes [10,16,17,25,26]. Reinhard et al. stated that contaminant removal is dependent on the chemical characteristics of both membrane and contaminants [25]. Further, molecular characteristics effecting solute sorption include water solubility and hydrogen bonding ability. Other features considered to influence transport include steric factors of branching and cross-sectional area. However, organic solutes characterized is often limited and consequently solute transport is poorly understood [10]. NF membranes vary in their rejection characteristics for smaller molecules such as chlorinated solvents and the relatively efficient removal of the halogenated one and two carbon volatiles may be due to the their poor H-bonding properties and due to their relatively higher halogenation's [10].

3.1. Effects of transmembrane pressures and feed concentrations

Total THM and its four compounds were chosen to evaluate the change of permeate concentrations over the course of filtration tests with varying transmembrane pressure levels. Fig. 2 illustrates the influence of transmembrane pressure on the rejection of THM over varying feed concentration with NF200 membrane. THM rejections increased a little for 20 and 80 μg/l feed concentration. After 80 μg/l feed concentration, the THM

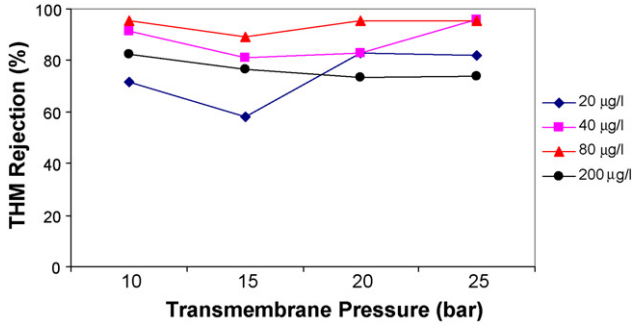

Fig. 2. Influence of transmembrane pressure on rejection of TTHM over varying feed concentration with NF200 membrane.

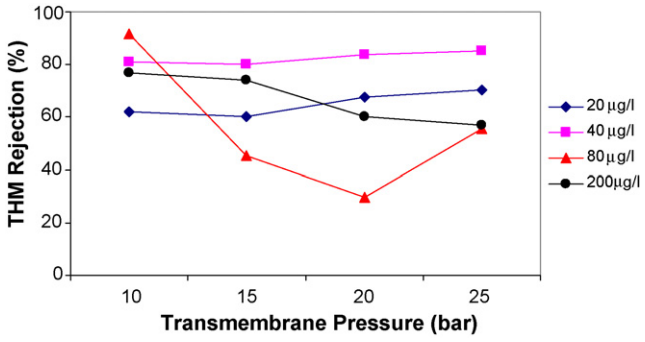

Fig. 3. Influence of transmembrane pressure on rejection of TTHM over varying feed concentration with DS5 membrane.

rejection efficiency does not show a substantial concentration changes. Besides, in general maximum THM rejection ratios were obtained at 10 bar transmembrane pressure under all feed concentrations. Interestingly, the amount of THM removal with NF200 membrane was 95% for 80 μg/l feed concentration at all pressure levels except 15 bar (Fig. 2). However, the rejection degree determined at higher pressures for NF200 membrane was reduced to a level of 74%. Besides, the THM rejection efficiency of DS5 membrane was found to be lower than those of NF200 membrane. In contrast to NF 200 membrane, the lowest THM removal rates at 80 μg/l of feed concentration were obtained at 20 bar operating pressure for DS5 membrane (Fig. 3). Further, as summarized in Fig. 2, rejection of THM decreased after pressure of 10 bar in terms of 20 and 80 μg/l of feed concentrations with DS5 membrane.

As reported by other researchers that, rejection of DBP compounds was influenced by compound physical chemical properties (e.g., molecular size, solubility, diffusivity, polarity, hydrophobicity, and charge), membrane properties (e.g., permeability, pore size, hydrophobicity, and charge), and membrane operating conditions (e.g., flux, transmembrane pressure, and recovery) [10,16,17,25–27], both diffusion and partitioning are highly related to physical and chemical properties of compounds and membranes.

On the other hand, there is clear explanation for the decline in THM rejection efficiency associated with increasing transmembrane pressure from 10 to 25 bar with two membranes studied.

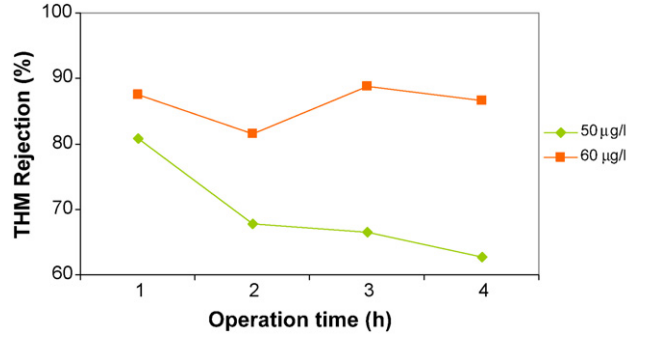

Fig. 4. Influence of operation time on rejection of TTHM over varying feed concentration with DS5 membrane.

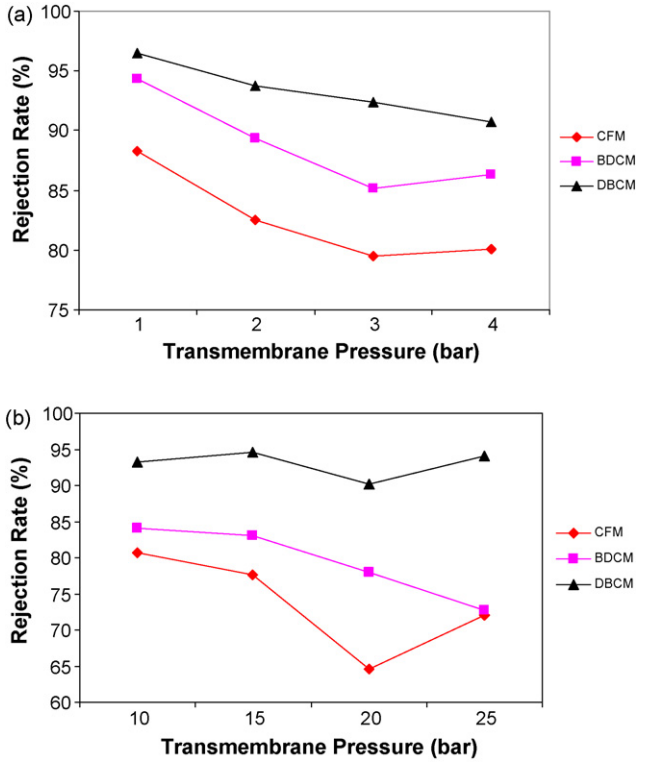

Fig. 5. Influence of transmembrane pressure on permeate concentration of THM species with 200 μg/l feed concentration for (a) NF200 membrane and (b) DS5 membrane.

Based on these results, it can be suggested that the THM rejection efficiency of both membranes was affected by THM compounds molecular sizes and pK_a values. In addition to steric obstacle, Kiso et al. determined the hydrophobicity of compounds quantified as *n*-octanol/water partition coefficient (K_{ow}) as another key parameter for rejection [28–30].

On the other hand the use of NF for the removal of THMs has not been investigated by researchers extensively yet. Rejections of total THM for two different influent concentrations are shown in Fig. 4. Rejections of THM decreased as filtration proceeded with 50 μg/l, but for 60 μg/l, the rejection trend exhibits smooth line with increasing operation period.

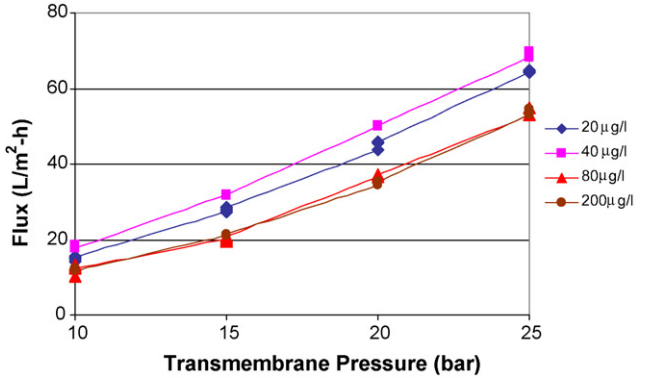

Fig. 6. Influence of the feed concentration over varying transmembrane pressure on the permeate flux for THM with NF200 membrane.

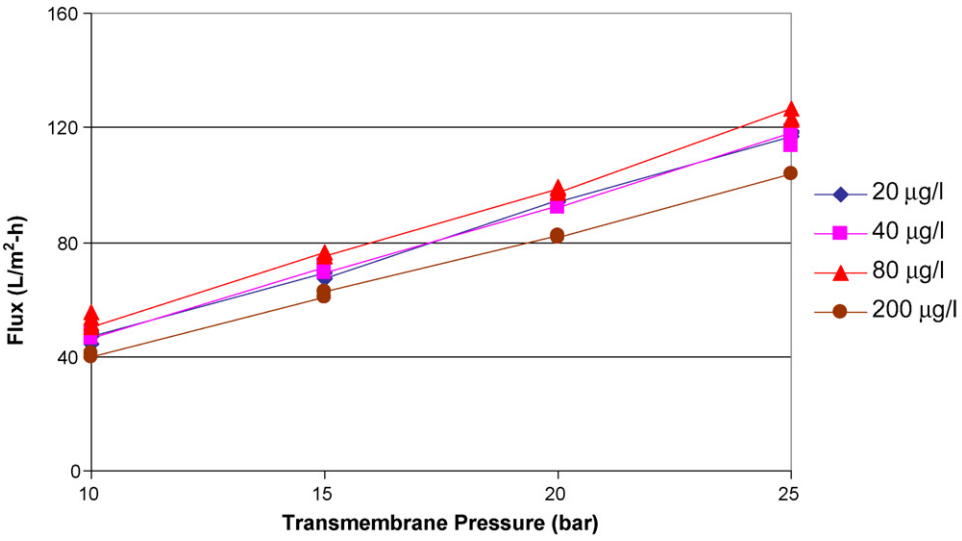

Fig. 7. Influence of the feed concentration over varying transmembrane pressure on the permeate flux for THM with DS5 membrane.

3.2. Effects of THM speciation on rejection capacity

Fig. 5 shows the rejection performances of three species of THM by two NF membranes: NF200 and DS5. The THM species are chloroform (CFM), bromodichloromethane (BDCM), and dibromochloromethane (DBCM). Since chlorination of raw water did not result in formation of bromoform (BFM), hence, three species of THM were taken into account. The NF tests were conducted under four different transmembrane pressures. The NF200 membrane was so effective in removing the THM compounds (Fig. 5). It was depicted in Fig. 5 that the rejection efficiencies of CFM, BDCM, and DBCM were found to be 88, 94, and 96, respectively. As observed with other studies, the removal efficiencies of NF200 were increased with increasing MW of THM species (Table 2) [10,17]. The results show that NF200 membrane was so effective in removing CFM, BDCM, and DBCM. Brominated THMs also did show the same removal trend as the chlorinated THMs. As bromine atoms replace the chlorine atoms, greatly increasing the MW, resulted in higher removal efficiency. The higher removal efficiency of DBCM was attributed to higher MW and brominating characteristics. For the DS5 membrane, Fig. 5 illustrates the removals of CFM, BDCM, and DBCM to be 81, 84, and 93, respectively. The rejection rates of THM compounds were decreased with increased transmembrane pressures. As was the case with NF200 membrane, DS5 rejection efficiency of THM species was found to be higher. Increasing operating pressures have a negative effect on rejection.

3.3. Flux characteristics of THM solution

On the other hand, the NF200 and DS5 membrane was tested to determine the influence of THM feed concentration over varying membrane pressure on the permeate flux. Experiments were carried out with solutions containing THM having a concentration of 20, 40, 80, and 200 µg/l. Besides, transmembrane pressure of 10–25 bar were applied. As shown in Figs. 6 and 7, the values of permeate flux were increased with increasing pressure. As stated by Koyuncu et al. [31], permeate flux is directly related to the feed pressure and osmotic pressure differences [31]. Jiraratananon et al. [32] stated that osmotic pressure differences increase with increasing contaminant concentration and so permeate flux decreases [32]. The evaluation of permeate flux observed in this study is shown in Figs. 6 and 7. Interestingly, permeate flux was observed not to decrease with increasing THM concentration. These results may be explained by low level of THM concentration in feed solution.

4. Conclusions

Nanofiltration is a technology that has potential for use in drinking water treatment. The conclusions that can be drawn from the results of this experimental investigation are as follows:

- THM rejection efficiency did not show a substantial concentration change with increasing transmembrane pressures. Besides, in general maximum THM rejection ratios were obtained at 10 bar transmembrane pressure under all feed concentrations.
- The experimental results show that both NF membranes were effective in removing CFM, BDCM, and DBCM. Further, brominated THM compounds also were removed significantly as was the case with chlorinated THMs. As bromine atoms replace the chlorine atoms, greatly increasing the MW, resulted in higher removal efficiency. The higher removal efficiency of DBCM was attributed to higher MW and brominating characteristics.
- On the other hand, permeate flux was observed not to decrease with increasing THM concentration. These results may be explained by low level of THM concentration in feed solution.

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Glossary

BDCM: bromodichloromethane
BFM: bromoform
CFM: chloroform
DBCM: dibromochloromethane
DBPs: disinfection by-products
D/DBP: disinfectant/disinfection by-product
DIUF: distilled ultra filtered
EU: European Union
GC/μECD: gas chromatograph/micro electron capture detector
GAC: granular activated carbon
HAA: haloacetic acids
MGD: million gallons per day
MW: molecular weight
NF: nanofiltration
NOM: natural organic matter
SUVA: specific ultraviolet absorbance
SOCs: synthetic organic chemicals
TLW: Terkos Lake water
THMFP: THM formation potential
TTHM: total THM
THM: trihalomethanes
USA: United States of America