



UV treatment for the removal of bromate formed during ozonation of groundwater. Influence of the oxidation process on the removal efficiency

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ABSTRACT

The presence of bromide ions in waters treated with ozone can lead to bromate ions and brominated compounds formation, potentially carcinogenic. This poses a need to remove the bromates through other treatment, as UV. In this work the effects of the water composition, and the corresponding variation due to oxidation, on the UV efficiency for the reduction of bromate to bromide ions in groundwater (GW) were investigated. For this purpose an experimental apparatus was specifically designed and experimental tests with ozone and with a UV lamp 6 W were performed. The experimental results show that the reduction of the bromates to bromides by UV is max 41% in GW, depending on ozone dose and initial bromate concentration. The experimental data were fitted with a first order model and the kinetic constant determined. The effects of the ozone and UV treatment on others groundwater components, as TOC and inorganic nitrogen, were taken into account and the corresponding influence on the bromate removal efficiency was evaluated. To this aim, comparative tests with UV in purified water (PW)(reagent grade) and GW spiked with controlled amount of bromates were performed. It was verified that the removal efficiency of bromate by UV increases with increasing of ammonia concentration, but decreases with increasing of TOC and when intermediate bromide species, produced by ozonation, are presents.

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1. Introduction

The bromide ions (Br^-) are present in natural waters at different concentrations, between 10 and 3000 ($\mu\text{g/L}$) and in seawater at much higher concentrations (67 mg/L) [1,2].

Their presence is due to natural and anthropogenic processes. Some natural processes are the infiltration of sea water and the washing action of the water on the rocks. Among the anthropogenic sources can be mentioned the waste products by extraction of potassium or coal, the fertilizers and pesticides [3], and the 1,2-dibromoethane used as scavenger in leaded fuel [4].

The use of Advanced Oxidation Processes (AOPs) to treat polluted waters, containing bromides, can lead to undesired formation of harmful brominated organic compounds [5] and

bromate ions (BrO_3^-) [6]. Since the 90s, the International Agency for the Research on Cancer (IARC) has classified bromate as a potentially carcinogenic and the European law has established a concentration limit in drinking water of 10 ($\mu\text{g/L}$).

Among the AOPs, the use of ozone, alone or combined with other oxidant agents and/or catalyst, is a well-established technology [7–10]. In removal processes that involve ozone, the oxidation can occur via ozone itself, via hydroxyl radical, $\cdot\text{OH}$, or via a combination of both. Besides, since ozone and $\cdot\text{OH}$ radicals are not selective, together with the beneficial effects, the oxidation of the other compounds contained in water can produce undesired by-products, such as the case of bromate [11]. The paths to bromate formation may occur simultaneously or in sequence, giving rise to an extremely complicated and highly no-linear reactions system [12]. The amount of bromates formed during the ozonation process of bromides is also affected by several parameters such as pH, the initial concentration of bromides, temperature, ozone dose and the overall water composition. Therefore the concentration of the

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bromates at end of an ozonation treatment could exceed the law limits and consequently the water would require further treatments.

There are many studies about methods to remove the bromate from water [13–17]. Some authors studied a semiconductor photocatalyst, platinized titanium dioxide (Pt/TiO_2) that can be used to promote the reduction of bromate ions to bromides and oxygen through irradiation with UV at 254 nm. The Pt/TiO_2 was used as powder dispersion or as a thin film in a flow reactor [18]. The bromate decomposition with low-pressure mercury vapor lamps was studied in buffer-free and buffered Milli-Q water by following the fate of bromine species, namely BrO_3^- and Br^- [19]. It was confirmed that BrO_3^- was converted to Br^- and that the decay followed a pseudo-first-order kinetic; moreover it was verified that the rate constant increased with photon flux and it was independent by initial concentration of bromate and initial pH [19]. Siddiqui and Amy [6] observed a high reduction of bromate in drinking water pretreated with ozone, with increasing of UV intensity. In fact, an increasing of the intensity from 850 to 2500 ($\mu\text{W}/\text{cm}^2$) caused a growing of bromate removal efficiency from 3% to 50%, with an initial bromate concentration in the range 50–100 ($\mu\text{g}/\text{L}$). Another study on drinking water with an initial bromate concentration of 50 ($\mu\text{g}/\text{L}$), reported a removal efficiency of 40% as a result of a UV dose of 2500 (J/m^2) [20]. It was also founded that the presence of high concentrations of organic matter and nitrates can decrease the effects of UV radiation due to the formation of radical species that can re-oxidize the bromides to bromates [21].

Nevertheless, there is still a lack of information about the reduction of bromate to bromide by UV in ozonised GW, and in particular about the role of the variation in the matrix composition due to the oxidation.

In this study the bromate formation from bromides contained in GW, due to ozonation, and its removal by UV were investigated. To this aim, experimental tests with GW containing bromide were performed. In particular the GW was first ozonized and then irradiated with UV. Since the ozone dose plays a critical role, its effect on the bromate formation was evaluated at six different levels in the range 8.43–50 (mg/L). Tests of UV treatments, without pre-ozonation, on PW (reagent grade) and GW spiked with bromates were performed in order to assess the roles of the water matrix composition and of its variation due to oxidation respectively.

An experimental apparatus was specifically designed in order to study the two processes, oxidation and reduction, separately and avoiding interferences.

The experimental data obtained from reduction of bromates were fitted with first order model and the rate constant was evaluated experimentally.

Finally the groundwater samples were analysed throughout the treatments, in order to verify the effects on some natural chemical components. Particular emphasis was given to the organic matter and inorganic nitrogen.

2. Materials and methods

2.1. Groundwater (GW) and purified water (PW)

The samples of the GW were drawn by an electric submersible pump from a 40 m deep well, in ENEA's Research Centre Portici located near Naples (Italy). The GW and PW were characterized before being used in the experimental tests.

The GW was filtered through a 0.45 μm Whatman membrane and analysed for its principal chemical constituents by methods described in Section 2.4. The results are reported in Table 1. The PW was obtained with a Water Purification System equipped with MilliQ gradient A10 plus and Elix 3 in series by Millipore S.A.S.,

Table 1

Means values of main chemicals present in GW used for experimental tests. The GW analysis was repeated at the beginning of each experiment (*) by Ion Chromatography, (**) by Flow Injection Analysis, (***) by TOC Analysis.

Parameters	Concentration (mg/L)	
Br^- (*)	0.153	± 0.008
BrO_3^- (*)	<0.1	
NO_3^- (*)	124.44	± 8.9
NO_2^- (**)	0.0156	± 0.002
NH_3 (***)	0.006	± 0.001
TOC (***)	0.79	± 0.05
PO_4^{3-} (**)	0.28	± 0.015
F^- (*)	3.30	± 0.19
Cl^- (*)	107.20	± 6.3
SO_4^{2-} (*)	151.71	± 7.3
pH	7.00	± 0.2

France. The PW quality is consistent with purity standards described by ASTM®, ISO® 3696 and CLSI® norms and has the following principal characteristics: 18 $\text{M}\Omega$ resistivity, Total Organic Carbon (TOC) < 5 ($\mu\text{g}/\text{L}$), bacteria < 0.1 (cfu/ml), Cl^- < 1 ($\mu\text{g}/\text{L}$), Na^+ < 1 ($\mu\text{g}/\text{L}$).

2.2. Experimental apparatus

Two sections composed the experimental apparatus: ozone and UV treatments (see Fig. 1).

The ozonation section (Fig. 1a) was designed as a semi-batch plant: with continuous flow for the gas and batch for the liquid. This section and the ozone production were described in previous paper, as well as the determination of liquid holdup for each flow gas [10].

After the ozonation treatment, the column was connected to the UV circuit through a valve system. The two sections have in common the reactor column (BC in Fig. 1).

The UV section (Fig. 1b) is designed as a batch plant, where a pump recirculates the liquid through the circuit formed by a column and a UV lamp. The lamp is coated by a quartz cylinder and is located in a 304 stainless steel tube, with an inlet and an outlet, of 26 cm of length and 5 cm of diameter. The UV lamp used is a Low Pressure Mercury Lamp (LPML), Philips mod. TUV 6 W, emitting a monochromatic radiation at the wavelength of 254 nm. A thermocouple placed at the exit of the UV reactor allows monitoring the water temperature.

2.3. Experimental procedure

In Table 2 the operating conditions of the experimental tests are reported.

First experimental tests with ozone and UV were performed on GW (Tests 1–6).

The values of different ozone doses (total amount of oxidizing agent by solution volume), were calculated by:

$$D = \frac{C_{\text{O}_3\text{in}} Q_G t}{V_L} \quad (1)$$

where $t = 15$ min is the total duration of a single test, Q_G is the feed gas (oxygen) flow to the ozone generator, $C_{\text{O}_3\text{in}}$ is the ozone concentration in the feed gas and V_L is the volume of treated liquid. For each feed gas flow value set, two values of doses were obtained through ozone generator adjustments.

In each test, the liquid was first exposed to O_3 for 15 min (at doses equal to 8.43, 10.3, 13.3, 31.6, 36.2, 50.0 mg/L). Subsequently, after 45 min from the end of ozonation, water samples were treated in UV circuit for 180 min. The interval time after the end of ozonation and before irradiation was required to allow the

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