A short review on bromate in wastewater – its formation, removal and mitigation

Sofia Högstrand, PhD student at the Department of Chemical Engineering, Lund University
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Abstract

The observation of the negative impacts of pharmaceutical residues and other organic micropollutants (OMPs) on ecosystems and drinking water production has in recent years led to an increase of measures to reduce the spread of these substances to the environment. For this purpose, ozonation and activated carbon-based processes have been deemed viable within wastewater treatment. One issue with the ozonation process is the formation of disinfection by-products (DBPs) such as bromate. Bromate is formed when bromide is present in the water and the ozone dose is high enough. Removal of bromate has been found difficult, yet there are three main measures with which formation can be mitigated: 1) reducing bromide concentration, 2) lowering the ozone dose, and 3) adding hydrogen peroxide or altering pH. In Lidköping municipality, Sweden, there are plans on implementing an ozonation step when constructing a new wastewater treatment plant (WWTP). However, initial samplings indicate high concentrations of bromide in the incoming water. An investigation of major sources is planned, and mitigation measures should be taken if these values are confirmed.

1 Introduction

The occurrence of pharmaceuticals and other organic micropollutants (OMPs) in wastewater effluents have in recent years become a global point of concern. As of today, mainly two methods have been successfully developed and considered viable in full-scale treatment to remove or degrade these substances at the wastewater treatment plants (WWTPs); adsorption via activated carbon and oxidation by ozonation (Eggen *et al.*, 2014).

Switzerland is the first, and still only, country to adopt national regulations on emissions of OMPs and has begun to implement full-scale tertiary treatment aiming at reducing and/or removing these substances. Germany has also begun to actualize full-scale treatment complying instead to regional recommendations of OMP removal. In Sweden, Nykvarnsverket in Linköping is the first WWTP to include full-scale ozonation in tertiary treatment as a means of OMP reduction and was constructed in 2017. As of 2018, the Swedish Environmental Protection Agency (Naturvårdsverket) launched a campaign to address this issue by granting financial resources to several municipalities and water- and wastewater corporations in Sweden. With this investment, they both aim at research regarding and constructing facilities for OMP reduction (Naturvårdsverket, 2018).

Lidköping municipality, frequently confused with the previously mentioned Linköping, is today in the initial stage of constructing a brand new WWTP. The old plant is underdimensioned and its location is desired for other purposes as it is located right at the shore of lake Vänern. Therefore, a new WWTP will be built outside of town thus opening up for implementation of various techniques and new designs without having to make compromises due to already existing facilities. Among other processes, such as enhanced biological phosphorus removal (EBPR) with subsequent struvite precipitation and disc filters to remove microplastics; an ozonation step for OMP reduction is planned.

However, it has been discovered that the incoming water to the existing WWTP (and thus highly likely also to the future one) contains high levels of bromide. Bromide occurs naturally in e.g. marine environments and may thus be spread by seaborne aerosols but as Lidköping is located far from the ocean the source is more probably anthropogenic (Sturges and Harrison, 1986). Bromide itself would not pose such a problem as it has a rather low toxicity. Nevertheless, when treating water with high levels of bromide with ozonation there is a great risk of obtaining brominated disinfection by-products (Br-DBPs) or, which may be of even greater concern, bromate (see further elaboration on the toxicology in sections 2.3 and 3.1).

To confirm these measurements, source mapping will be initialized to further identify the source of the bromide. In the meantime, it would be valuable to evaluate possible effects if the influent to the new WWTP would contain similar concentrations of bromide as the existing one. The aim of this study is therefore to generate an initial overview of the scientific field with comparisons to the case of Lidköping.

1.1 Research questions

For this purpose, the below stated questions will be investigated.

- What causes bromate formation?
- What can be done to remove or reduce bromate?
- How can bromate formation be mitigated?
- What measures would be applicable for the case of Lidköping?

1.2 Method

This will be a brief overview of existing literature, thus the method used is searching relevant articles in databases such as Web of Science. A few articles have also been recommended by supervisors or colleagues in the LIWE LIFE project. Furthermore, information regarding the planned WWTP in Lidköping such as preliminary process schemes, descriptions and sample data have been provided by colleagues within the LIWE LIFE project.

2 Bromide (Br^-)

Information regarding sources, occurrence and toxicology of bromide is given in this section.

2.1 Sources

As mentioned above, bromide exists naturally in the oceans and is spread by atmospheric aerosols. Yet, the concentrations of marine bromide diminishes further inland and instead anthropogenic causes are more probable explanations of bromide occurrence (Sturges and Harrison, 1986). There are several anthropogenic uses of bromide of which brominated flame retardants, biocides, the pulp and paper industry, treatment of industrial cooling water but also residential pool water treatment are a few examples (Vainikka and Hupa, 2012). Thus, chemical industries, special waste industry and municipal waste incinerators are the most significant point sources of bromide to WWTPs (Soltermann *et al.*, 2016).

2.2 Occurrence

During a recent study in Switzerland on the origin and occurrence of bromide in Swiss surface waters it was observed that in rural areas the bromide concentrations are rather low (<20 µg/L) whereas in areas of greater anthropogenic influence the concentrations could amount up to 55 µg/L (Soltermann *et al.*, 2016).

Regarding concentrations of bromide in WWTP effluents, the same study concluded that for the major part of the studied WWTPs, these concentrations were also rather low (<100 µg/L). Nevertheless, at WWTPs with major bromide sources in their catchments (such as chemical and special waste industry or waste incineration plants) the effluent concentrations were distinctly higher (>400 µg/L) (Soltermann *et al.*, 2016).

2.3 Toxicology

There are no current recommendations regarding wastewater effluent concentrations of neither bromide nor bromate or brominated disinfection by-products. Bromide is a substance of low toxicity with a drinking water concentration recommendation of 0.4 mg/kg body weight. This is said to correspond to 24 mg/person or 6 mg/L for someone who weighs 60 kg or 2 mg/L for a 10 kg child who drinks two respectively one litre of water per day (WHO, 2009).

3 Bromate (BrO_3^-)

In this section, the toxicology, formation and removal of bromate are presented.

3.1 Toxicology

Bromate is considered a potential carcinogen with a drinking water concentration limit of 10 µg/L (WHO, 2005; European Council, 1998; US-EPA, 2006). Regarding ecotoxicological limits Hutchinson *et al*. (1997) proposed a maximum level for bromate of 3 mg/L. Another, more recently, proposed environmental quality standard for both short- and long-term exposure of bromate is 50 µg/L (Oekotoxzentrum Eawag-EPFL, 2015).

3.2 Bromate formation

Formation of bromate during ozonation treatment occurs as the present bromide is oxidized via reactions with both ozone and OH-radicals (von Gunten, 2003). Apart from bromate, during oxidation, other transformation products such as brominated disinfection by-products (Br-DBPs), are also formed (Hollender *et al.*, 2009). However, Chon *et al*. (2015) showed that for specific ozone doses (SOD) of < 0.25 g O_3/g DOC there were no pronounced formation of bromate as the ozone instead was reacting with dissolved organic matter. Soltermann *et al*. (2016) stated that for SODs > ~0.4 g O_3/g DOC, almost linear correlations to bromate formation was obtained. In experimental studies it has been observed that at the bromide levels tested (16-31 µg/L) the bromate in the effluent were maximum 7.5 µg/L and thus still below the drinking water standards of 10 µg/L also for the highest tested ozone dose of 1.16 g O_3/g DOC (Hollender *et al.*, 2009; Zimmermann *et al.*, 2011).

3.2.1 A Swiss example

The regulations in Switzerland regarding OMP removal states that 80% of twelve indicator OMPs need to be removed over the treatment plant. For this purpose, required ozone doses ranges normally between 0.4-0.6 g O_3/g DOC which would (for a typical wastewater) render ≤ 3% of molar bromate yield (Soltermann *et al.*, 2016). Bromate yield is defined as produced bromate per concentration of bromide in the incoming wastewater.

The first WWTP in Switzerland to implement full-scale ozonation was Neugut WWTP. Doses between 0.35-0.97 g O_3/g DOC where evaluated in order to find an optimum dose that would comply with the regulations on OMP removal. Based on a ≥ 80% removal of the above mentioned indicator substances, a SOD of 0.55 g O_3/g DOC was concluded to be necessary (Bourgin *et al.*, 2018). The incoming wastewater contains bromide levels comparable to common Swiss municipal WWTPs (Soltermann *et al.*, 2016); ranging from 35 to 85 µg/L with an average of 60 µg/L and measured maximum of 150 µg/L. When ozone doses were increased so was the bromate formation; with a bromide level of 60 µg/L and an ozone dose of 0.97 g O_3/g DOC the bromate concentration in the effluent reached 5.7 µg/L. However, at the average bromide level (60 µg/L) and recommended ozone dose (0.55 g O_3/g DOC) the bromate formation was 1.8 µg/L which corresponds to a bromate yield of 3%. Furthermore, due to dilution in the recipient the actual concentrations there are normally < 0.18 µg/L which is clearly below both the previously mentioned drinking water standard and the proposed ecotoxicological limits (Bourgin *et al.*, 2018).

3.3 Bromate removal

Many organic DBPs are to some extent biodegradable and due to this reason, a biological treatment following ozonation is often incorporated into the treatment train. However, what is of special concern regarding bromate is its incapability of being biodegraded in such a subsequent biological treatment (von Gunten, 2003). It has also been observed by Zimmermann *et al*. (2011) that bromate concentration was not reduced by aerobic sand filtration. On the other hand, it has been noticed that combining ozonation with a subsequent biological activated carbon (BAC) treatment step has efficiently reduced both Br-DBPs and bromate concentrations when using empty bed contact times of 15 minutes (Chuang and Mitch, 2017). The authors concluded that a possible reason for the removal of the bromate could be attributed to the low dissolved oxygen (DO) concentrations and they referred to Kirisits *et al*. (2001). These authors, in turn, investigated the factors influencing biological bromate removal (using BAC filters) from surface- and groundwater after ozonation. They established that the removal was enhanced at lower DO and nitrate concentrations as well as at lower pH-values. However, when Gerrity *et al*. (2011) investigated the effect of combining O_3/H_2O_2 with BAC they did not observe any biological reduction of bromate despite the presence of anaerobic microbes. Therefore, as bromate is difficult to remove from the process it is of greatest importance instead to mitigate its formation (von Gunten, 2018).

3.4 Bromate mitigation

There may be difficulties removing bromate from WWTP effluent but there are means that can be taken in order to mitigate its formation. Soltermann *et al*. (2016) suggest three main measures: 1) reducing bromide concentration, 2) lowering the ozone dose and 3) adding hydrogen peroxide. A further discussion on the subject was published a year later containing a more elaborated analysis (Soltermann *et al.*, 2017). A collection of their thoughts and findings is presented in this section.

3.4.1 Reducing bromide concentration

Bromate formation is directly linked to concentration of bromide in the incoming water. Thus, by reducing this concentration, either through point source removal or bromide removal from the wastewater itself, bromate formation could be avoided. Several techniques are available for the latter (Watson *et al.*, 2012), but due to various reasons (such as high costs, energy consumption or ineffectiveness) they are not regarded as suitable solutions in full scale wastewater treatment (Soltermann *et al.*, 2017).

The other option is to remove point sources of bromide to the WWTP. Soltermann *et al*. (2016) stated that for most WWTPs in Switzerland, the incoming concentrations of bromide are so low that for an ozone dose of 0.4-0.6 g O_3/g DOC the bromate concentrations in the effluent would be below the drinking water standard of 10 µg/L after ozonation. On the other hand, in WWTPs with incoming water containing effluents from a major point source of bromide, the concentrations could reach up to a few milligrams per litre. Therefore, removing these waters from the inflow to the WWTP could prove to be an efficient measure to reduce bromate formation.

Removing point sources of bromide to the WWTP could be done by installing a separate treatment at these bromide-rich locations (see examples in section *2.1 Sources*). After treatment the water is discharged directly into surface waters if all the legal criteria regarding other polluting factors such as heavy metals, DOC concentration and pH are fulfilled (Soltermann *et al.*, 2017). It is also important to consider that due to frequently varying bromide concentrations in the effluent from point sources (Soltermann *et al.*, 2016), long term monitoring of concentrations and loads at these sources, in the sewer systems as well as in the WWTPs is vital in order to estimate the actual effects of each respective point source (Soltermann *et al.*, 2017).

3.4.2 Lowered ozone dose

As mentioned in section *3.3 Bromate removal*, implementation of BAC may not have a direct effect on bromate removal though indirectly it could as it allows for a lower ozone dose (and thus less formation of bromate) but still removes micropollutants. This combination of both ozonation and activated carbon could prove to remove even a greater range of pollutants than using them separately (Soltermann *et al.*, 2017). Another option might be to interrupt ozonation completely during times of bromide peak loads. This is, however, mainly suitable with longer peaks (range of days) and not with shorter ones (range of hours). Furthermore, discontinuation of the ozonation results in less micropollutant removal and therefore affects the reason as to why ozonation was implemented in the first place (Soltermann *et al.*, 2017).

3.4.3 Addition of hydrogen peroxide or altering pH

The molecular reactions of bromate formation was investigated by von Gunten and Oliveras (1998) and they concluded that there are two major pathways; one with $HOBr/OBr^-$ as intermediate and the other with a BrO-radical as intermediate. The former pathway, they concluded, is affected by addition of hydrogen peroxide (H_2O_2) which would result in a lowered bromate formation. However, as there is another pathway the extent to which bromate formation is mitigated could vary depending on yet other parameters. The authors meant that this might explain the varying results of H_2O_2 implementation regarding bromate formation mitigation that has been presented previously in literature.

The implementation of the advanced oxidation process (AOP) where ozone and hydroperoxide are combined has continued to be studied since. Wert *et al*. (2007) noted that addition resulted in a lowered formation of bromate, yet their conclusion when comparing multiple factors was that conventional ozonation would be recommended over the O_3/H_2O_2 AOP. Soltermann *et al*. (2016) suggest it as one possible solution of bromate formation mitigation. However, as the authors elaborate the subject further, they conclude that even though addition of H_2O_2 has proven successful in drinking water treatment, the method is of minor importance within wastewater treatment due to the dual pathway system (Soltermann *et al.*, 2017). Furthermore, even though bromate formation may be mitigated by addition of H_2O_2, it is important to consider that the effect of H_2O_2 addition depends on the composition of the wastewater. Besides, addition of H_2O_2 might also inhibit the reduction of micropollutants (Soltermann *et al.*, 2017).

The other major pathway is affected by OH-radicals and reducing these by lowering the pH has been shown to be effective within drinking water treatment (Pinkernell and von Gunten, 2001). In wastewater treatment, on the other hand, the objective is to reduce the broad range of micropollutants as well as disinfect the water which renders both ozone and OH-radicals vital. This strategy is thus not suitable here (Soltermann *et al.*, 2017).

3.4.4 Ozonation suitability

There are many parameters to consider when determining if ozonation is the best available method for OMP removal in treatment plants. It is therefore recommended to perform a thorough investigation of bromide occurrence and bromate formation potential before implementing ozonation at a WWTP (Soltermann *et al.*, 2016). Schindler Wildhaber *et al.* (2015) developed a test procedure for facilitating such an evaluation. The methodology includes evaluations of 1) ozone stability in the wastewater matrix, 2) OMP removal efficiency, 3) formation of DBPs, and 4) assessment of toxicity via bioassays.

Even though bromide in itself is not considered to negatively affect the recipient ecosystems at the current expected concentration levels, it may, however, affect the possibility of implementing oxidative treatment. This means that if the potential of bromate formation is high enough there could be cause for restricting the implementation of an ozonation step. (Soltermann *et al.*, 2016).

4 Ängen WWTP

Lidköping municipality is located on the southern shore of lake Vänern. The current WWTP is situated in the centre of town, at the harbour. The new treatment plant will instead be placed outside, southeast of town. As mentioned in the introduction, the plant will contain processes such as EBPR with struvite precipitation and disc filters for a polishing step to remove e.g. microplastics. Between these processes, there will be an ozonation step for OMP removal and disinfection of the wastewater. In this section, the ozonation configuration and the incoming bromide concentration will be discussed.

4.1 Process configuration

The new WWTP in Lidköping municipality, Ängen WWTP, is still in its planning stage. Therefore, the process configuration presented in this section might be subject to further alterations and may thus not be representative of the final plant.

The ozonation facility will consist of two ozone generators, one contact tank where the ozone will be mixed with the incoming water, and then there will be a subsequent biological treatment. The contact tank will have a volume of 300 m^3 and a recommended hydraulic retention time (HRT) of 10-25 minutes to make sure that all ozone has time to react. The maximum flow through the contact tank is 1120 m^3/h, which corresponds to 80% of the maximum flow in the biological treatment. The flow regulates the ozone dose which is designed as 5.6 g O_3/g m^3 and corresponds to 0.7 g O_3/g DOC for a DOC-content of 8 mg/L (based on German and Swiss experiences). Should flow and DOC-content not correlate in such a way, there are possibilities of adjustments later on. Maximum ozone consumption, i.e.

at a flow of 1120 m^3/h, would amount to 6.3 kg/h. It is argued that in order to avoid bromate formation it is better to have lower ozone doses. Instead the contact time will be longer so that OMP reduction is still ensured.

The subsequent biological treatment will consist of a moving bed biofilm reactor (MBBR). Due to high oxygen levels in the effluent water from the contact tank, there was an initial plan to by-pass a small part of the incoming water to the ozonation directly to the MBBR in order to allow for a more rapid consumption of the oxygen. This would be done so that post-denitrification could take place if deemed necessary. However, there has been a discussion concluding that this post-denitrification would probably not be necessary as the biological treatment prior to the ozonation step should be able to reach sufficiently low nitrogen levels. It would instead be of greater benefit regarding primarily hygienisation but also micropollutant removal to let all water go through the ozonation step. Therefore, this by-pass will probably not be included in the finished WWTP and the MBBR will instead be focussed on aerobic degradation of transformation products.

4.2 Incoming bromide concentrations

The new treatment plant will be designed for 61 000 people equivalents (PE) of which 25 000 PE will come from a liquor production industry. The effluent from this industry has a high but rather varying concentration of DOC and suspended solids (SS). It is, however, assumed that this effluent will be subjected to pre-treatment before entering the municipal sewage network. Regarding possible sources of bromide, there is a waste incineration plant for district heating in the town and it is suspected that this might be at least partly responsible for the high bromide levels in the incoming water.

Long term monitoring of the wastewater is required in order to be able to draw any conclusions. Despite this, initial samplings may indicate that the bromide concentrations going out of the biological treatment in the current WWTP is ~300 µg/L which is several times higher than the Swiss average of < 50 µg/L for WWTPs without major sources of bromide. Instead, these values are more in correlation with the bromide concentrations from such a WWTP with high loads of bromide in the influent (~400 - 50 000 µg/L) (Soltermann *et al.*, 2016). A few samples taken right at the effluent from the liquor industry show bromide levels of ~70 µg/L. Further down the sewage network where this wastewater is mixed with effluent from the incineration plant the one sample taken here shows high levels of bromide (1400 µg/L). Note that this is only one sample and more measurements are required. Nevertheless, this indicates influence from the incineration plant rather than the liquor industry.

Therefore, if these numbers are confirmed in the aforementioned source mapping investigation, bromate mitigating actions will be necessary. One way this could be facilitated is through point source removal. If it is indeed the waste incineration plant that is the sole or main source, then this could prove to be the most efficient measure. If there are many different or diffuse sources, the option of implementing a subsequent BAC filtration step could instead be evaluated to allow for a lower ozone dose and thus less formation of bromate.

It may also be of interest to perform an investigation such as the one presented by Schindler Wildhaber *et al*. (2015) to evaluate the suitability of ozonation at the WWTP. However, it is important to remember that there are no regulations regarding bromate concentrations in wastewater effluent. The results in literature are therefore mainly compared with drinking water standards. Thus, it could be questioned whether this limit is relevant for wastewater implementation, or if it is too strict. The proposed environmental quality standard for long- and short-term exposure is higher and could perhaps be more reasonable for recipient analysis. Furthermore, the dilution of wastewater effluent into the receiving water is of great importance as it could render a wastewater effluent highly loaded with bromate still environmentally acceptable if the recipient is large enough. Also vice versa is true; a wastewater effluent with a lower concentration of bromate might be considered too harmful if the recipient is of smaller size.

5 Conclusions

To conclude, the answers to the questions put up in section 1.1, would from this initial overview be:

- Bromate formation is caused by oxidative treatment of bromide-containing waters. For ozonation, the bromate formation reaction is impacted both by ozone and OH-radicals in a dual pathway system. Doses of < 0.25 g O_3/g DOC renders little bromate but increasing doses > ~0.4 g O_3/g DOC shows linear correlations to bromate concentration.
- Removing or reducing bromate has been showed to be difficult. There are indications that biological removal through BAC filters may be successful within drinking water treatment. However, regarding its use for wastewater treatment the research is contradictory and rather inconclusive.
- Bromate formation can be mitigated through three main measures; 1) Reducing bromide concentration (mainly through point source removal), 2) Lowering the ozone dose (by implementing a subsequent activated carbon treatment) and 3) Adding hydrogen peroxide (though this is probably more relevant within drinking water treatment than in wastewater treatment) or altering pH (but that would negatively affect OMP removal).
- Initial sampling in Lidköping municipality indicates high levels of bromide in the incoming water to the WWTP. Thorough investigation of point sources is therefore already planned. If these values are confirmed, mitigation measures such as point source removal or combining the ozonation with BAC filtration in order to lower the ozone dose could be possible solutions. It is, however, also important to consider what effluent concentrations could be acceptable based on emission goals and recipient sensitivity and dilution.

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7 Abbreviations

BAC	Biological Activated Carbon	OMP	Organic MicroPollutants
DBP	Disinfection By-Product	SOD	Specific Ozone Dose
EBPR	Enhanced Biological Phosphorus Removal	WWTP	WasteWater Treatment Plant

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