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Biostability of surface waters are potentially deteriorated as a result of partial oxidation of natural organic matter under UV/H₂O₂ treatment that is applied for the removal of micropollutants. This research highlights the impact of using coagulation to remove natural organic matter prior to the UV/H₂O₂ process by utilizing a robust and rapid technique for gauging the changes in the biostability.

1 Pretreatment of natural organic matter to control biological stability

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5 Abstract

6 Application of UV/H₂O₂ process for degradation of micropollutants in surface waters could
7 deteriorate the biological stability of the treated water. This is because of the partial oxidation of
8 natural organic matter under the applied UV/H₂O₂ conditions that in turn leads to increase in the
9 assimilable organic carbon (AOC). To address this issue, Alum coagulation was investigated as a
10 NOM pretreatment alternative prior to the UV/H₂O₂ process in order to improve the treatment
11 efficacy and the water quality. A recently developed technique was utilized to rapidly assess the
12 AOC of treated water at various stages. Alum was effective at removing a substantial portion of
13 large to medium size organic molecules that led to a considerable reduction in AOC. However, the
14 fractions not removed by coagulation showed to promote some levels of bacterial regrowth after
15 undergoing subsequent UV/H₂O₂ treatment. That said, Alum pretreatment was found to be an
16 effective strategy for reducing the formation of AOC by 14 to 40 % depending on the water used
17 and UV dose applied. Findings of this study are of interest for utilities that already have coagulation
18 in use and seek to comply with more upcoming stringent regulations by incorporating advanced
19 oxidation processes (e.g., UV/H₂O₂) in their treatment train.

20 **Key-words:** UV/H₂O₂, Natural Organic Matter, Advanced Oxidation Processes, Assimilable
21 Organic Carbon, Biological Stability, Coagulation

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23 1 Introduction

24 Advanced oxidation process (AOP e.g., UV/H₂O₂) is one of the most effective alternatives for the
25 elimination of organic and trace-level micropollutants in water¹⁻⁴. However, efficacy of UV/H₂O₂
26 for the abatement of target contaminants is diminished by the presence of natural organic matter
27 (NOM) in water. During UV based AOPs (e.g., UV/H₂O₂ treatment), NOM shields UV and
28 scavenges OH radicals (HO[•]) generated within the process, thereby reducing the efficacy of
29 treatment. The UV dose and H₂O₂ concentration applied are designed to degrade micropollutants
30 and therefore NOM is only partially oxidized and is broken down into smaller more biodegradable
31 molecules^{5,6}. These smaller organics are reported to be the one of the major contributors to the
32 bacterial re-growth and biofilm formation within distribution systems^{7,8} and are usually referred to
33 as assimilable organic carbon (AOC). Therefore, removing NOM (i.e., completely or partially) via a
34 pretreatment process could potentially enhance the performance of UV/H₂O₂ at removing
35 micropollutants and also improve the biostability of treated water.

36 Several processes have been proposed and examined in the literature for the removal of NOM under
37 various water loading and qualities⁹⁻²¹. Among the options proposed, coagulation process such as
38 the use of Alum, Ferric Chloride, PACl is well established and commonly applied in large scale
39 applications^{11,12,16,22}. Moreover, it could serve as a viable pretreatment prior to UV/H₂O₂ process,
40 because of the recognized ability of coagulants (e.g., Alum, Ferric Chloride, PACl) to remove a
41 considerable portion of medium-high molecular weight NOM, and its relatively straightforward
42 operation²³⁻²⁶. Therefore, the main objective of this research was to gauge the impact of
43 coagulation (e.g., Alum) on degradation of NOM and its subsequent effect on the biological
44 stability (i.e., AOC) of UV/H₂O₂ treated water.

45 Two natural water sources were selected and preliminary coagulation tests were conducted to assess
46 the optimum Alum dose for NOM removal. Changes in physiochemical properties (such as UV₂₅₄,
47 total organic carbon (TOC) content, and NOM molecular weight distribution) for raw, Alum
48 treated, UV/H₂O₂ treated, and Alum-UV/H₂O₂ treated water samples were carefully assessed and

49 monitored. A recently developed AOC bioassay using flow cytometry that was previously modified
50 for UV/H₂O₂ treated waters ²⁷ was utilized to quantify AOC in all stages ^{28,29}. Findings of this
51 research are potentially of interest for those utilities that already use coagulation process (e.g.,
52 Alum, Ferric Chloride, PACl) and hence, could readily implement advanced oxidation processes
53 (e.g., UV/H₂O₂) in their treatment train to meet future more-stringent guidelines.

54 2 Materials and Methods

55 2.1 Source water characteristics

56 Raw waters for this study were collected from Bowen Island (BI, UV₂₅₄=0.183 cm⁻¹, TOC~4.81
57 ppm, pH~ 6.7) and Capilano Reservoir (CR, TOC~1.45 ppm, UV₂₅₄=0.061 cm⁻¹, pH~ 6.3) both
58 located in British Columbia, Canada.

59 2.2 UV/H₂O₂ treatment

60 A collimated beam set-up utilizing a low pressure (LP) high output amalgam lamp was used to
61 conduct UV/H₂O₂ treatment experiments. The selection of LP UV lamp was because of its wider
62 application due to lower cost and energy use, as well as lower potential of forming hazardous by-
63 products ^{1,3,30-33}. Water samples were initially filtered (0.45 μm) and spiked to initial H₂O₂
64 concentration of ~10 ppm (H₂O₂ 30%, Fischer Scientific). Next, samples were irradiated to the
65 desired UV fluence (i.e., up to 2000 mJ/cm²) as determined by chemical actinometry ³⁴.
66 Experimental conditions (e.g., UV dose and H₂O₂ concentration) were selected based on previous
67 researches ^{1,5,6,15,30,35,36} and detailed description of the collimated beam set-up and the experimental
68 procedure is described elsewhere ^{6,35}.

69 2.3 Coagulation-UV/H₂O₂ treatment

70 Alum (Al₂(SO₄)₃·18H₂O, ACS reagent +98%, Sigma Aldrich) was used as the coagulation reagent
71 for NOM removal. Jar test applied involved two minutes of rapid mixing at 150 rpm followed by 30
72 minutes of slow mixing at 60 rpm. Floccs formed were then allowed to settle for an hour.
73 Preliminary results indicated 5 and 15 ppm as the most effective doses of Alum in terms of TOC

74 and UV₂₅₄ reduction for CR and BI waters, respectively (data not shown). The Alum-treated
75 samples were then filtered (pre-rinsed 0.45 µm filter) to remove any potential particles/floc (that
76 may not have settled) and then underwent UV/H₂O₂ treatments as described earlier.

77 2.4 Analytical Methods

78 Several water quality parameters (e.g., UV₂₅₄, total organic carbon (TOC), and molecular weight
79 distribution) were monitored to study the fate of NOM during the treatment. The concentration of
80 H₂O₂ was measured using the triiodide method³⁷. A UV-Vis spectrophotometer (Shimadzu UV-
81 Mini 1240, cell path length of 1 cm) was used to conduct all the spectrophotometric measurements
82 (e.g., UV₂₅₄, [H₂O₂]). A Shimadzu TOC-VCPH analyser was used to measure the total and
83 dissolved organic carbon (TOC, DOC) content of water. High performance size exclusion
84 chromatography (HPSEC) technique was used to analyse the apparent molecular weight (AMW)
85 distribution of NOM with a similar procedure described by Sarathy and Mohseni (2007)⁵. A
86 WATERS 2695 HPLC system equipped with a 2998 photodiode detector, set to detection at 260
87 nm, served as the instrument for HPSEC analysis. Calibration curve correlating the AMW to
88 retention time was obtained from polysulfonate standards (7 kDa PSS7K, 4 kDa PSS4K, 2 kDa
89 PSS2K, American Polymer Standards Corporation), acetone, and benzoic acid (10 ppm)⁵.

90 2.5 AOC bioassay

91 To eliminate any potential for cross contamination, all glassware materials were thoroughly
92 cleansed and baked at 550 °C for 5 hrs. All other tools (e.g., pipette tips) were autoclaved to ensure
93 minimizing the risk of contamination. Residual H₂O₂ (after UV/H₂O₂ treatment) has been shown to
94 have detrimental effect on the growth of microorganisms and AOC bioassay²⁷; hence, it was
95 removed using Catalase from bovine liver immobilized on a polymeric substrate (SEPABEAD®,
96 Resindion, Italy). Details on the preparation and validation of this analytical method is described
97 elsewhere^{27,38}. Then, AOC bioassay was performed according to the protocol proposed by Hammes
98 and Egli (2005)²⁸ with some modifications as described elsewhere²⁷. In brief, a natural indigenous
99 inoculum, made from the source water, was utilized instead of the conventionally used pure strains

100 i.e., P17 and NOX. Ultrapure (Milli-Q) water spiked with various levels of sodium acetate (>
101 99.99%), all filtered through 0.22 μm , served as the control throughout all experiments. Samples
102 quenched from H_2O_2 were seeded with the natural inoculum and incubated for 72 hrs at 30 $^\circ\text{C}$ in
103 amber vials. Flow cytometry in combination with fluorescence cell staining was used to count the
104 cells grown, providing more accurate and reliable data. A flow cytometer unit (FACS-Callibur
105 System Becton, Dickinson and Company) located in the Biomedical Research Centre at the
106 University of British Columbia (UBC) was used to carry out the cell analysis. Detailed procedure
107 on cell enumeration and AOC data analysis is extensively described elsewhere ²⁷.

108 **3 Results and Discussion**

109 3.1 Changes in physicochemical characteristics

110 Alum coagulation alone resulted in $\sim 50\%$ and 73% reduction of TOC and UV_{254} , respectively, for
111 CR water. Similar impact was observed on BI water as the TOC and UV_{254} were reduced by 63%
112 and 80% , respectively. Control experiments involved the treatment of raw water with $\text{UV}/\text{H}_2\text{O}_2$
113 with 10 ppm peroxide and UV fluences of up to $2000 \text{ mJ}/\text{cm}^2$ (in the absence of Alum). Table 1
114 shows the TOC and UV_{254} reductions for the raw and alum-treated waters irradiated under the UV
115 fluence of $2000 \text{ mJ}/\text{cm}^2$. As a result of the extensive $\text{UV}/\text{H}_2\text{O}_2$ treatment (i.e., $2000 \text{ mJ}/\text{cm}^2$, control
116 experiments), UV_{254} was reduced between $35\text{-}67\%$ while considerably lower TOC reductions 9-
117 26% was recorded for the waters tested. The larger fractional decrease of chromophoric NOM
118 (CNOM, represented by UV_{254}) in both treatment cases (i.e., $\text{UV}/\text{H}_2\text{O}_2$, $\text{UV}/\text{H}_2\text{O}_2$ after coagulation)
119 implies the preference of generated OH radicals to react with UV_{254} absorbing compounds and
120 partial oxidation of the organic matter. Similar findings has also been reported by other researchers
121 ^{5,35}.

122 Moreover, Alum is known to be effective in removing large to medium range molecular weight
123 organics ^{11,12,39}. Therefore, downstream $\text{UV}/\text{H}_2\text{O}_2$ treatment was expected to give larger fractional
124 CNOM reductions due to the absence of larger organics that preferentially react with HO^\bullet ^{5,35,40}. It is

125 noteworthy that lower absolute reductions in UV_{254} and TOC were observed for the Alum treated
 126 water samples as a result of the UV/H₂O₂ treatment (Table 1). This could be mainly explained by
 127 the pseudo-first order reaction of TOC and CNOM with OH radicals generated during the UV/H₂O₂
 128 process³⁵. That is with lower initial organic matter lower reaction rate is expected; however higher
 129 UV transmittance would result in higher amount of generated OH radicals thereby compensating for
 130 the organic concentration term³⁵.

131 **Table 1: Reductions in UV_{254} and TOC of raw and Alum-treated natural waters during**
 132 **UV/H₂O₂ process (values in parenthesis indicate absolute reduction).**

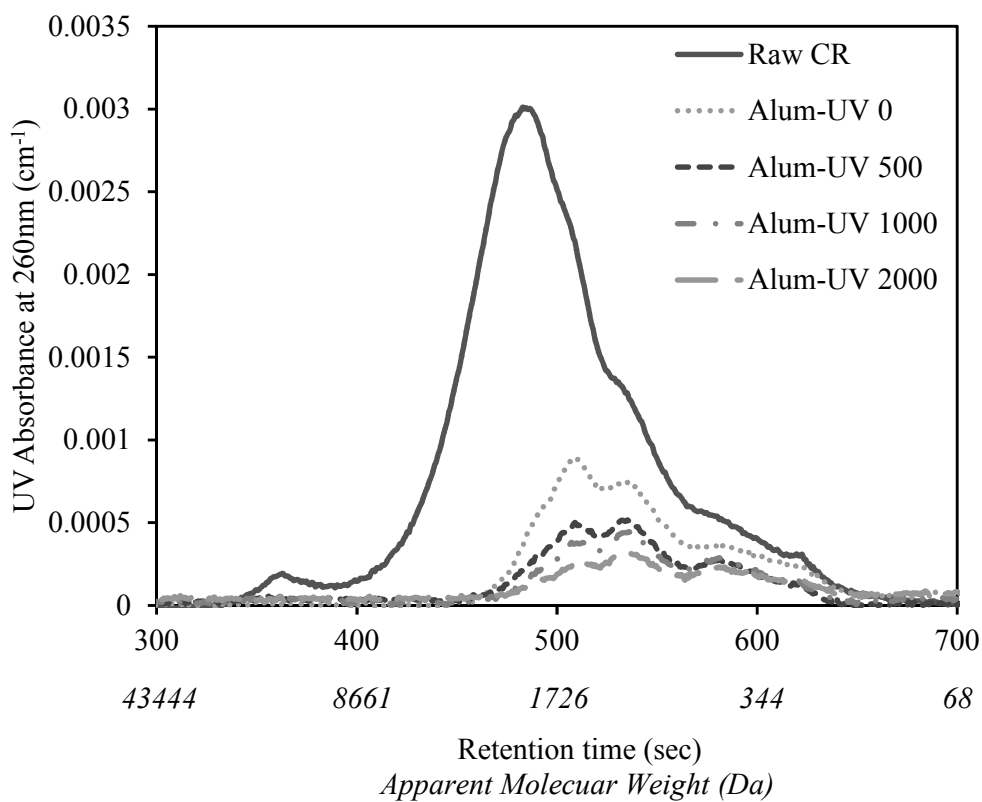
	CR water		BI water	
	Raw water (control)	Alum-treated	Raw water (control)	Alum-treated
UV_{254}	52.5% (0.032)	66.7% (0.010)	34.4% (0.063)	52.9% (0.018)
TOC	25.9% (0.384)	18.0% (0.131)	12.1% (0.58)	8.7% (0.151)

133

134 Figure 1 shows the changes in the molecular size distribution of alum-pretreated CR water that has
 135 undergone various UV/H₂O₂ treatment extents. As demonstrated, the use of Alum alone (i.e., Alum-
 136 UV 0) was effective at removing a substantial portion of NOM, mainly larger molecular weight
 137 fractions. Consistent with the literature, Alum showed to preferentially remove organics of high to
 138 medium molecular weight range¹¹. Application of UV/H₂O₂ treatment after Alum resulted in
 139 further decrease in the AMW of UV absorbing NOM, up to the UV fluence of 500 mJ/cm².
 140 However, no considerable change was observed by extending the UV dose beyond 500 mJ/cm².

141 Similar observation was recorded for the changes in the NOM molecular weight distribution of BI
 142 water as shown in Figure 2. As illustrated, Alum coagulation eliminated the first large eluting peak,
 143 often associated with colloidal organic matter, as well as considerable portions of other molecular
 144 weights mainly from large to medium weight organic molecules (i.e., > 500 Da). However, further

145 downstream UV/H₂O₂ treatment resulted in little reduction in the remainder of the chromophoric
146 organic molecules.

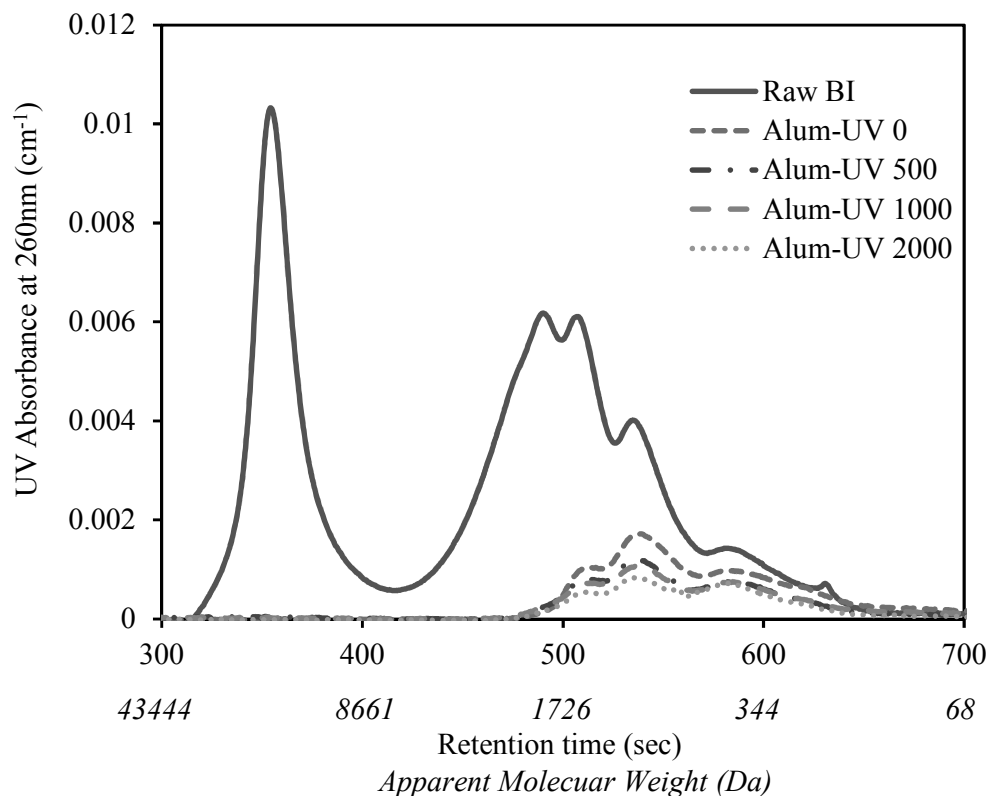


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148

Figure 1: Impact of UV/H₂O₂ treatment on AMW of Alum-treated CR NOM.

149



150

151 **Figure 2: Impact of UV/H₂O₂ treatment on AMW of Alum-treated BI NOM.**

152 3.2 Impact on biostability and AOC

153 Coagulation noticeably reduced the AOC of CR and BI waters. The reduction was more significant
 154 for BI water (88.2% compared to 83% for CR), likely attributed to the elimination of the large
 155 single eluting peak corresponding to high molecular size organics in the HPSEC chromatogram
 156 (Figure 2). This is also consistent with the greater TOC and UV₂₅₄ reductions observed for BI water
 157 (compared to CR water) as it underwent the alum coagulation.

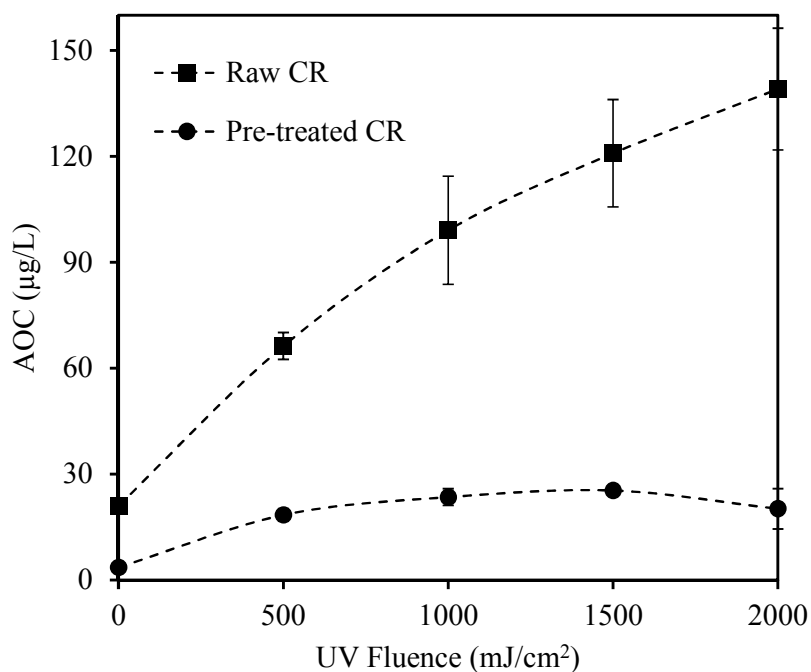
158 Figures 3 and 4 compare the AOC profiles of raw and Alum-treated for CR and BI waters under
 159 various UV doses. As shown, UV/H₂O₂ advanced oxidation resulted in significant increase in AOC
 160 for raw waters. This is because of the partial oxidation and breakdown of (larger) organic molecules
 161 (i.e., into smaller ones) as a result of reaction with OH radicals. Figures 3 and 4 also depict the
 162 ability of the organics not removed via coagulation to promote bacterial growth even though their

163 observed AOC values were very low. Interestingly, the AOC of the Alum-treated waters still
164 increased (and then plateaued) under the UV/H₂O₂ treatment, supporting the fact that further
165 structural breakdown of NOM molecules (even though mostly of lower molecular weight nature)
166 took place. That said, using the HPSEC technique was not sufficient to capture all the changes in
167 the molecular structure of NOM. Nonetheless, as expected the absolute increase in AOC was
168 noticeably lower for the waters pretreated with Alum. This was because Alum removed a
169 considerable portion of large-medium organic molecules which are the most susceptible ones
170 towards reaction with OH radical. As a result of Alum treatment, the assimilable percentage of
171 NOM (i.e., AOC/TOC×100) decreased from 1.42% and 1.56% (in raw waters) to 0.49% and 0.51%
172 for alum-treated CR and BI waters, respectively. This confirms a considerably lower assimilable
173 fraction remained after coagulation. Observations made here are also in agreement with the
174 observation of Chong Soh et al. (2008)¹², who also found the remaining NOM fractions after
175 coagulation were able to support bacterial regrowth.

176 Both Alum-treated waters showed greater fractional AOC increase (7 and 12 folds, respectively,
177 under UV/H₂O₂ treatment) in comparison with the raw waters (5 and 3.5 folds increase,
178 respectively). Also, as previously shown in Table 1, greater fractional UV₂₅₄ reduction was
179 observed for the Alum-treated waters. Therefore, this can be mainly attributed to the more effective
180 interactions between OH radicals and smaller organic molecules (i.e., in the absence of high MW
181 OH scavenging dissolved organics), leading to higher enhancement in biodegradability of NOM.
182 That is, lower levels of organic matter would result in smaller UV₂₅₄ absorbance consequently
183 leading to higher UV absorption rate by H₂O₂^{35,41}. Therefore, higher fractional AOC increase of the
184 pretreated waters would be expected as less shielding and scavenging effects of NOM exists. As a
185 result, more effective number of interactions/reactions between OH radical and organic matter
186 would be expected^{6,35}.

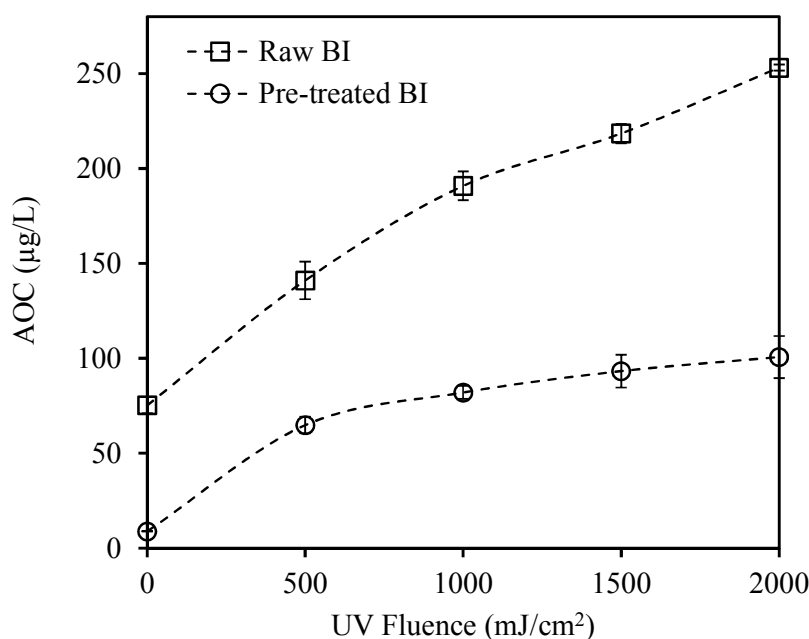
187 The behaviour of the AOC profiles for pretreated CR and BI waters over the course of UV/H₂O₂
188 treatment is also noteworthy (Figures 3 and 4). After the UV fluence of 1000 mJ/cm², the AOC

189 profiles begin to plateau, indicating a possible equilibrium with respect to the formation of smaller
190 (biodegradable) organics and their subsequent degradation with OH radicals. Moreover, AOC of the
191 pretreated CR started to decrease slightly after the UV fluence of 1500 mJ/cm² (Figure 3). A likely
192 and plausible explanation is that, at this fluence, the degradation rate of organic molecules was
193 dominant and greater than the rate of formation; hence, an overall decrease in the amount of small
194 biodegradable organic molecules was observed.



195

196 **Figure 3: Impact of UV/H₂O₂ treatment on AOC of raw and Alum-treated CR water.**



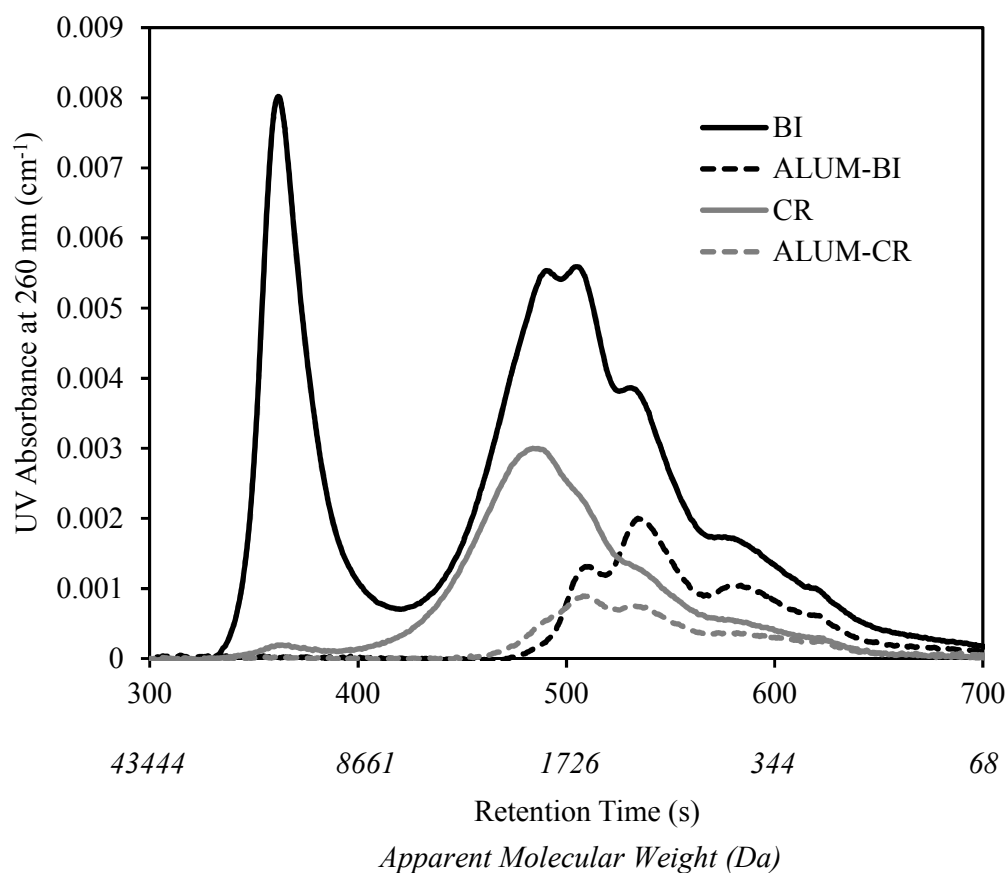
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198 **Figure 4: Impact of UV/H₂O₂ treatment on AOC of raw and Alum-treated BI water.**

199 Moreover, the AOC of the pretreated CR water increased by about 7 times at UV fluence of 2000
200 mJ/cm², however, the AOC of pretreated BI water showed increases of about 12 times from its
201 initial value with the same level of the treatment (Figures 3 and 4). This could be attributed to the
202 higher organic content (i.e., TOC) as well as the nature of NOM in the source water. That is,
203 differences in the nature and characteristics of NOM in BI and CR waters resulted in different
204 behaviours and responses with respect to their reaction with OH radicals and consequently
205 biodegradability increase under identical AOP treatment. Also, Alum treated BI water contained
206 higher amount of organic molecules in the range of low-medium molecular weights (AMW < 1000
207 Da) in comparison with the alum treated CR water (Figure 5). As a result, the remaining organics
208 in BI water were likely more biodegradable to start with and underwent further partial oxidation,
209 leading to greater amount and percentage of AOC generated during UV/H₂O₂ treatment (i.e., UV
210 fluence of 2000 mJ/cm²). Supporting either of these hypotheses require further investigations using
211 more analytical techniques such as liquid chromatography equipped with organic carbon detection,
212 use of isolation techniques (e.g., nanofiltration), and also potentially evaluation of different source

213 waters. This will eventually lead to better understanding of the fate of NOM during various
214 treatments and the potentials for AOC formation.

215 Even though the AOC of the Alum treated water still increased over the course of UV/H₂O₂
216 process, it is important to note that the final AOC was comparable to that of the raw water (with no
217 treatment). This means that the combined treatment strategy did not significantly change the
218 biostability characteristic of the water. This is an important consideration because it indicates that
219 the application of combined Alum and UV/H₂O₂ may not deteriorate the biostability of the treated
220 water to level that it would require downstream biological treatment (e.g., biological activated
221 carbon). On the other hand, standalone UV/H₂O₂ with the resultant significant increase of AOC
222 could not be implemented without the application of downstream biological treatment to remove the
223 generated AOC.



224

225 **Figure 5: Comparison of AMW distribution of NOM for Raw and Alum-treated CR and BI**
226 **waters**

227 Data obtained from the standalone UV/H₂O₂ treatment along with those of the combined treatment
228 clearly indicated that the application of Alum significantly reduces the concentration of high
229 molecular weight NOM. Lower concentration of NOM, in particular from the higher molecular
230 weight fractions, leads to the more effective use of UV photons and lower competition for the OH
231 radicals in the water matrix. The lower scavenging of UV and OH radicals, in turn, leads to more
232 effective removal of target contaminants (i.e., micropollutants) which are the primary reason for the
233 application of UV-based AOPs. More importantly, this will help to conserve considerable amount
234 of electrical energy used for delivering the necessary UV fluence (to achieve the degradation of
235 target contaminants)³⁰.

236 One important note to consider is that the findings in here would be of interest for those facilities
237 that already have coagulation process in place. Otherwise, incorporating coagulation (coagulation,
238 flocculation, and sedimentation) for a new treatment plant may not be a feasible pretreatment
239 alternative since coagulation is a relatively expensive process.

240 **4 Conclusions**

241 Application of UV/H₂O₂ process to remove micropollutants has been shown to adversely affect the
242 biological stability of the treated water. This is because of the breakdown of large natural organic
243 matter molecules (as a result of reaction with HO[•] and UV) into smaller ones that can be readily
244 consumed by bacteria (i.e., AOC). To address this issue, Alum coagulation was used as a NOM
245 pretreatment alternative to eliminate medium-high molecular weight fractions upstream of the
246 oxidation process, hence mitigating the formation of smaller more biodegradable organic molecules
247 (i.e., AOC). A rapid and novel technique, modified to UV/H₂O₂ applications, was utilized to
248 monitor the AOC profile (i.e., concentration of biodegradable molecules) through coagulation and
249 UV/H₂O₂ treatment. AOC was reduced by 80-85 % as a result of Alum coagulation. However,
250 downstream UV/H₂O₂ process raised the amount of biodegradable organic molecules (AOC), with
251 the extent of the AOC increase dependant on the nature of the NOM and the treatment conditions
252 (e.g., treatment time).

253 Overall, application of a pretreatment process (e.g., Alum) prior to the UV/H₂O₂ can potentially
254 reduce the risk of deteriorating the biostability while saving a considerable amount of electrical
255 energy to achieve the same level of target contaminants removal.

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