**SUPPLEMENTAL MATERIAL**

**S3. RCT Analysis**

The removal of 1,4-dioxane from ozone and ozone/ultrasound was found to be strongly dependent on the radicals formed. The RCT approach (Elovitz & von Gunten, 1999) was used to study the ozone consumption and mass transfer properties in the continuous flow reactor. The RCT concept relies on the assumption that the target contaminant, 1,4-dioxane in this case, has a low reactivity with ozone and a very high reactivity with hydroxyl radical. Dioxane meets this requirement, since the rate constant with ozone is kO3/1,4-dioxane = 0.32 M-1s-1 (Hoigne & Bader, 1983) and that with ∙OH is kOHˑ/1,4-dioxane = 2.4 x 109 M-1s-1 (Suthersan & Payne, 2004). Equation 4 shows the rate of dioxane concentration as a function of ozone concentration.

|  |  |
| --- | --- |
|  | (1) |

The RCT term was then calculated using the influent and effluent concentrations of 1,4-dioxane, the rate constant for hydroxyl radical/1,4-dioxane reaction, and the time integrated ozone concentration. Given the continuous flow nature of this study, the aqueous ozone concentrations were measured at the inlet and outlet of the reactor. The time integrated ozone concentration was estimated as the mean of the inlet and outlet concentrations, multiplied by the treatment time. The mean RCT value determined from Figure 9 was approximately 1.33x10-8 for all ozone, ultrasound, pH, and pressure conditions tested using the drinking water matrix. This value is similar to the average RCT reported for twelve natural surface and ground waters in Switzerland, which was determined to be 1.56 (±1.6) x 10-8 (Elovitz, von Gunten, & Kaiser, 2000). The value obtained in this study was also comparable to the RCT value of para-chlorobenzoic acid (1.4 x 10-8), a compound with has been studied as a hydroxyl radical probe (Elovitz & von Gunten, 1999) (Pi, Schumacher, & Jekel, 2005). This is expected since 1,4-dioxane has also been used as a hydroxyl radical probe itself (Kishimoto & Sugimura, 2010) (Kitamura, Kishimoto, Okura, & Otsu, 2011) (Kishimoto, et al., 2008).

In Figure S3, the mean RCT and 95th percentile confidence intervals are given for a set of experimental results across a wide variety of process conditions: pH, pressure, and ultrasound. These results suggest that the hydroxyl radical yield was similar across all pressure, pH, ultrasound, and ozone dose conditions tested. The outliers in figure S3 are those experiments where the influent drinking water was spiked with 10mM and 100mM of bicarbonate and those experiments done using deionized water in lieu of drinking water. This is expected, since bicarbonate scavenges hydroxyl radicals, leaving fewer to react with 1,4-dioxane, and also because the bicarbonate has been shown to be a significant RCT-determining water characteristic (Elovitz, von Gunten, & Kaiser, 2000). The RCT findings suggests that the improved 1,4-dioxane removal noted in the combined ozone/ultrasound tests was almost entirely due to sonication-driven increased ozone consumption, rather than any phenomena related solely to ultrasound.



Figure S1: Matrix Effects on Ultrasound, Ozone, and combined Ozone/Ultrasound



Figure S2: Effect of bicarbonate as radical scavenger (6.5 min retention time)



Figure S3: RCT Line 95% confidence intervals for 1,4-Dioxane removal from spiked drinking water (no added HCO3)using O3 and combined O3/US



Figure S4: Model validation at [O3]aq = 1mg/L