Supplementary Information

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S.1 Complete uptake model for the aqueous reaction of I⁻ with O_3 at high I⁻ concentrations

At high I⁻ concentrations, as uptake coefficients approach values near 1, it is possible that the surface reaction of ozone with iodide is not completely parallel to the analogous aqueous-phase reaction. In this case, the complete uptake model would be described by the following equation:[1-3]

$$\frac{1}{\gamma^{I-}} = \frac{1}{\alpha_s^{I-}} + \cfrac{1}{\cfrac{1}{\alpha_{s,I-} \cdot \cfrac{k_{sol}^{I-}}{k_d^{I-}} + \cfrac{1}{\Gamma_{aq}^{I-}}} + \Gamma_s^{I-}}$$

(E1)

where most of the parameters have been defined in the main manuscript, and k_{sol}/k_d is the solvation to desorption ratio of O_3 in the aqueous system considered. At high I⁻ concentrations, this ratio is given by the following equation:[3, 4]

$$\frac{k_d}{k_{sol}} = \left(\frac{1}{\alpha_{aq}} - \frac{1}{\alpha_{s,I-}} \right) \alpha_{s,I-}$$

(E2)

Parameter α_{aq} is >0.002 and,[5] by definition, $\alpha_s > \alpha_{aq}$. For high I⁻ concentrations, $\alpha_s \approx \alpha_{s,I-}$. Therefore, k_d/k_{sol} is <500, but a lower limit of this ratio cannot be determined.

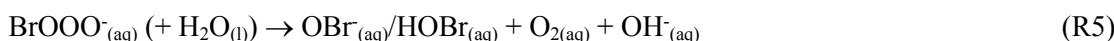### S.2. Parameters for the aqueous-phase reaction of O_3 with Br⁻ and with I⁻

S.2.1 Rate constant for the aqueous-phase of ozone with Br⁻ (k^{Br-})

The dependence of the total second-order rate constant for the reaction $Br^-_{(aq)}+O_{3(aq)}$ on pH has been proposed by Liu et al.[8]:

$$k'^{Br-} = \frac{k_1 \left(\frac{k_2}{k_{-1}}[H^+] + \frac{k_3}{k_{-1}} \right)}{1 + \frac{k_2}{k_{-1}}[H^+] + \frac{k_3}{k_{-1}}}$$

(E3)

where k_1, k_{-1}, k_2, k_3 are the kinetic constants of the reactions R3, -R3, R4, and R5, respectively:[8]

$$O_{3(aq)} + Br^-_{(aq)} \leftrightarrow BrOOO^-_{(aq)} \tag{R3}$$

$$BrOOO^-_{(aq)} + H^+_{(aq)} \rightarrow HOBr_{(aq)} + O_{2(aq)} \tag{R4}$$

$$BrOOO^-_{(aq)} (+ H_2O_{(l)}) \rightarrow OBr^-_{(aq)}/HOBr_{(aq)} + O_{2(aq)} + OH^-_{(aq)} \tag{R5}$$

The rate constants of these reactions are given as a function of temperature by the following equations:[9]

$$k_1 = 2.07 \cdot 10^{13} \cdot exp(-6840/T) \tag{E4}$$

$$k_2 = 1.50 \cdot 10^{39} \cdot exp(-25200/T) \tag{E5}$$

$$k_3 = 1.62 \cdot 10^{36} \cdot exp(- 24100/T) \tag{E6}$$

$$k_{-1} = 4.17 \cdot 10^{41} \cdot exp(- 26600/T) \tag{E7}$$

Artiglia et al.[9] used a slightly different correlation of the ozone solubility for the determinations of these rate constants than the one presently used, which is more recent (see section S.2.2). Therefore, the net rate constant will be obtained by multiplying equation E3 by a factor equal to the squared ratio of the solubility they used over the solubility used in the present work (see equation 8):

$$k^{Br-} = \frac{1.06 \; k_1 \left(\dfrac{k_2}{k_{-1}}[H^+] + \dfrac{k_3}{k_{-1}} \right)}{1 + \dfrac{k_2}{k_{-1}}[H^+] + \dfrac{k_3}{k_{-1}}}$$

(E8)

S.2.2 Ozone solubility

The ozone solubility in aerosols or solutions (s) depends on the temperature and on the identity and concentration of the present salt. It can be described by the Weisenberger-Schumpe equation:[6]

$$s = s_0 \cdot 10^{-\sum (h + h_{ion}) \cdot c_{ion}} \tag{E9}$$

where s_0 is the ozone solubility in pure water (expressed in liquid/gas concentration ratio) at a given temperature, c_{ion} is the concentration of a particular ion present, h_{ion} is the coefficient assigned to this ion, h is a coefficient that depends on the gaseous molecule (O_3) and on temperature, and the summation extends to all dissolved ions that are present. Rischbieter et al.[7] have obtained an empirical equation for the ozone solubility in pure water by evaluating previous determinations of this property. This equation is shown here in non-dimensional units:

$$s_0 = 10^{-5.12 + 1230/(T/K)} \cdot 8.31 \cdot 10^{-3} \, (T/K) \tag{E10}$$

The coefficient h for O_3 is given by the following equation:[7]

$$h = 3.96 \cdot 10^{-3} + 1.79 \cdot 10^{-3} (T/K - 298.15) \tag{E11}$$

where h is in M⁻¹ units. The coefficients h_{ion} for ion=Na^+, K^+, Br^-, I^-, Cl^- are shown in Table S1. Even though the correlation for the ozone solubility as a function of salt composition is only reliable up to an ionic strength of 1.5 M,[7] we will extend it to higher concentrations because, according to the authors' knowledge, there is no other available estimation method.

Table S1. Parameters to estimate gas solubilities in equation E10. Source: Weisenberger and Schumpe.[6]

Ion	h_{ion} (M⁻¹)
Na^+	0.107
K^+	$9.22 \cdot 10^{-2}$
Br^-	$2.69 \cdot 10^{-2}$
I^-	$4 \cdot 10^{-3}$
Cl^-	$3.2 \cdot 10^{-2}$

S.2.3 Diffusion coefficient of ozone in the aqueous phase

The diffusion coefficient of ozone in pure water (D_0) has been determined as a function of temperature:[10]

$$D_0 = 0.011 \; cm^2 \; s^{-1} \; exp(-1896 \; K/T) \tag{E12}$$

This empirical equation is valid for temperatures ≥ 10 °C, but we will extend it to slightly lower temperatures. The dependence of the diffusivity on salt concentration for aqueous media containing salts of I⁻, Br⁻, or Cl⁻ will be estimated using the Stokes-Einstein relation, by which the change in the diffusivity of a molecule in a solution relative to the diffusivity of the molecule in pure solvent is inversely proportional to the change in the viscosity of the solution relative to the pure solvent:[11]

$$D_{O_3} = D_0 \frac{\eta_0}{\eta} \tag{E13}$$

where D_{O3} is the ozone diffusivity in the solution, η is the viscosity of the solution, and η_0 is the viscosity of the pure solvent (in this case, water).

The viscosity of aqueous NaI and KI solutions of different concentrations have been obtained from Zaytsev and Aseyev[12], and the viscosity of NaBr, KBr, and NaCl solutions have been calculated following the method explained by Isono:[13]

$$\eta_{O_3} = \frac{hN_A}{V} exp\left(\frac{\Delta G_{vis}}{RT}\right) \tag{E14}$$

where h is the Planck's constant ($6.626 \cdot 10^{-34}$ m² kg s⁻¹), N_A is Avogadro's number ($6.022 \cdot 10^{23}$ molecule mol⁻¹), V is the mean molar volume of the solution, and ΔG_{vis} is the Gibbs energy of activation, which is given by:

$$\Delta G_{vis} = \Delta G_{vis}^0 + \beta_G x + \gamma_G x \tag{E15}$$

where ΔG^0_{vis} is the Gibbs energy of activation for pure water at 25 °C (9.17 kJ mol⁻¹)[13], x is the molar fraction of solute, and β_G and γ_G are parameters specific to the solute that are given by the following equations:

$$\beta_G = \beta_H - T\beta_S \tag{E16}$$

$$\gamma_G = \gamma_H - T\gamma_S \tag{E17}$$

where β_G, β_S, γ_G, and γ_S are parameters specific to the solute. The values of these parameters for NaBr, KBr, and NaCl solutions are shown in Table S.2. The mean molar volume of these solutions can be calculated from their density, which can be obtained from tables[12] as a function of molarity and temperature.

Table S.2. Parameters for equations E14-E15 to calculate the viscosity of NaCl, NaBr, and KBr solutions as a function of temperature and solute molar fraction. Source: Isono[13].

	β_H / 10² kJ mol⁻¹	γ_H / 10² kJ mol⁻¹	β_S / 10³ J K⁻¹ mol⁻¹	γ_S / 10³ J K⁻¹ mol⁻¹
NaCl	-0.56	6.16	-0.23	1.74
NaBr	-0.71	6.59	-0.26	1.81
KBr	-1.02	5.58	-0.32	1.54

The effect of presence of salts in the diffusivity is only significant for high salt concentrations. For example, at 25 °C the diffusivity decreases by about 5% and 40% for NaCl solutions of molarity 1 M and 5 M, respectively. Furthermore, small errors in the estimation of the diffusivity do not affect the total uptake very much because the uptake due to the aqueous-phase reaction is proportional only to $D^{1/2}$ (see equation 8). Unlike solutions containing Na⁺/K⁺/Br⁻/Cl⁻, the viscosity of NaI and KI solutions

decreases with concentration and therefore the aqueous-phase diffusion coefficient of gaseous molecules increases. However, the change is small and has therefore little effect on the uptake. For example, for a 3 M KI solution, the diffusivity is 9% higher than for pure water, leading to a 5% increase in the uptake by the aqueous-phase reaction.

S.3. Determination of the adsorption equilibrium constant of O_3 for Br^- solutions or aqueous particles (K_{O3}^{Br-})

S.3.1. Determination of K_{O3}^{Br-} at low temperature

The determination of K_{O3}^{Br-} at 0 °C can be calculated using the uptake coefficients obtained by Artiglia et al.[9] for constant $[Br^-]_{aq}$ and variable $[O_3]_{gs}$. From equation 14 it follows:

$$\Gamma_s^{Br-} = t\frac{K_{O_3}^{Br-}}{(1 + K_{O_3}^{Br-}[O_3]_{gs})}$$

(E18)

where t is a constant factor for constant $[Br^-]_{aq}$ and constant temperature that is equal to $4k_s^{Br-}[Br^-]_{surf}^{z+1}/\omega$. We have determined t and K_{O3}^{Br-} by minimizing the sum of the squared relative errors of the model-estimated uptake coefficients with respect to the experimental results[9] shown in Figure 2. The sum of errors for different values of K_{O3}^{Br-} is shown in Figure S1. As seen in this Figure, the optimal result is $K_{O3}^{Br-}=1.6\cdot10^{-13}$ cm^3 molecule^{-1}, but any value between $1.0\cdot10^{-13}$ and $1.7\cdot10^{-13}$ cm^3 molecule^{-1} leads to similar error. These results agree with the value of $1\cdot10^{-13}$ cm^3 molecule^{-1} that has been determined by Oldridge and Abbatt[14] using the same temperature, different NaBr concentration ($8.5\cdot10^{-3}$ M), and different composition of the solution (0.55 M NaCl + 0.01 M HCl), supporting that K_{O3}^{Br-} does not depend on the bromide nor chloride concentration. The comparison of model and experimental results is shown in Figure 2 of the main article.
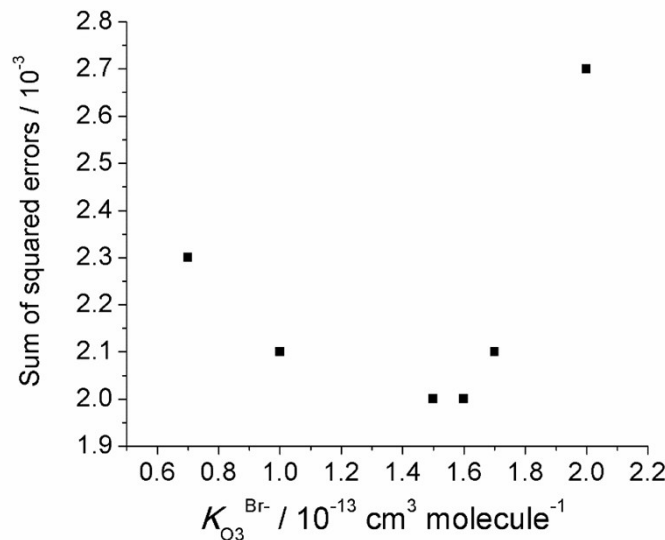

Figure S1. Sum of the squared relative errors of the model-estimated uptake coefficients relative to the uptake coefficients reported by Artiglia et al.[14] for [NaBr]=0.125 M and pH=5.6 at 0 °C for different assumed values of K_{O3}^{Br-}.

S.3.2 Determination of the temperature dependence of K_{O3}^{Br-}

The variation of $K_{O3}^{Br^-}$ with temperature can be estimated using the following equations:[15]

$$k_d^{Br^-} = A^{Br^-} \cdot exp\left(\frac{\Delta H_{ads}^{Br^-}}{RT}\right)$$

(E19)

$$K_{O_3}^{Br^-} = \frac{\alpha_{s,Br^-} \cdot \omega}{4N_{O_3}^{Br^-} \cdot k_d^{Br^-}}$$

(E20)

Substituting k_d from equation E19, $N_{O3}=[Br^-]_{surf}$ (see main text), and $\alpha_{s,Br^-}=p^{Br^-}\cdot[Br^-]_{surf}$ (see equation 17) into equation E20, the following relation between $K_{O3}^{Br^-}$ and temperature is obtained:

$$K_{O_3}^{Br^-} = \frac{p^{Br^-}\omega}{4A^{Br^-}}exp\left(-\frac{\Delta H_{ads}^{Br^-}}{RT}\right)$$

(E21)

In order to estimate the value of $\Delta H_{ads}^{Br^-}$, the value of $K_{O3}^{Br^-,0°C}$ estimated above as well as the following parameter values will be applied in equation E21:

- Parameter p^{Br^-} will be assumed to be equal to p^{I^-}, which has been estimated to be about $5\cdot10^{-3}$ M^{-1} in the main text and is approximately equivalent to $2\cdot10^{-16}$ cm^3 molecule^{-1}. Note that, according to equations E19-E21, $K_{O3}^{Br^-}$ does not depend on $[Br^-]_{surf}$ because both α_{s,Br^-} and $N_{O3}^{Br^-}$ are hypothesized to be proportional to $[Br^-]_{surf}$. The assumption that p^{I^-} equals to p^{Br^-} is adequate, as it has been found that $K_{O3}^{Br^-}$ is similar to $K_{O3}^{I^-}$ in the Model Results section.

- The mean thermal velocity of ozone (ω) at 0 °C equals $3.47\cdot10^4$ cm s^{-1}.

- The factor A^{Br^-} is estimated to be about $1\cdot10^{14}$ s^{-1} for molecules that adsorb chemically, such as O_3.[15]

Substituting these values in equation E21, the result is $\Delta H_{ads}^{Br^-}$=-67 kJ mol^{-1}. The mean thermal velocity of ozone (ω) varies only between $3.50\cdot10^4$ and $3.63\cdot10^4$ cm s^{-1} for the temperature range 1-25 °C. Therefore, an intermediate value of ω for these temperatures ($3.56\cdot10^4$ cm s^{-1}) will be used to obtain an equation for $K_{O3}^{Br^-}$ (cm^3 molecule^{-1}) for this temperature range. Introducing this value and the other parameter values indicated above in equation E21, the following equation is obtained:

$$K_{O_3}^{Br^-} = 2.0\cdot10^{-26}\cdot exp(8100\ K/T)$$

(E22)

There could be an error in the estimation of $K_{O3}^{Br^-}$ stemming from the uncertainty in parameter p^{Br^-}, constant A^{Br^-}, and the value of $K_{O3}^{Br^-}$ determined from Artiglia et al.'s data[9]. The largest source of error derives from A^{Br^-}, whose estimated limits of uncertainty are ($5\cdot10^{13}$, $5\cdot10^{14}$) s^{-1}. The effects of this uncertainty on the value of $K_{O3}^{Br^-,25°C}$ is 20%, which is quite low considering that this parameter can vary over orders of magnitude.

S.4 Model results for conditions used in previous studies

S.4.1 Error of the model-estimated ozone-dependence of the ozone uptake by Br⁻ solutions

Figure S2 shows the absolute error of the uptake coefficients estimated by the "preliminary model" for the interaction of ozone with a 0.125 M NaBr solution relative to the experimental results by Artiglia et al.[9] at different temperatures. These errors correspond to the uptake coefficients shown in Figure 2 in the main manuscript.
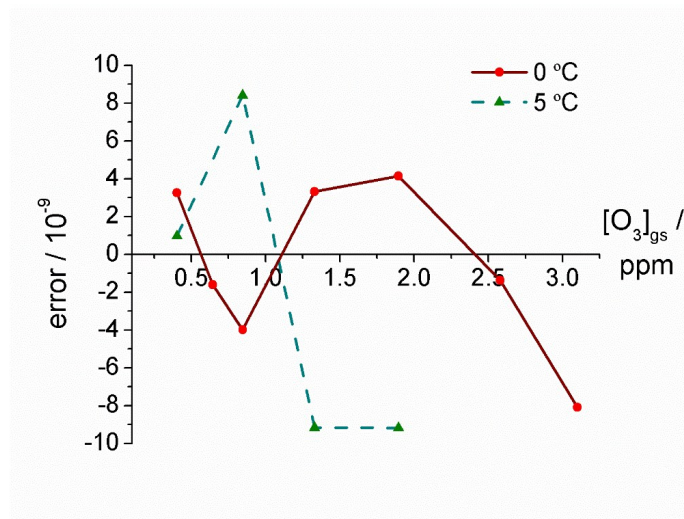
Figure S2. Absolute error of the uptake coefficients estimated by the "preliminary model" for the interaction of O_3 with a 0.125 M NaBr solution relative to the experimental results by Artiglia et al.[9] at different temperatures.

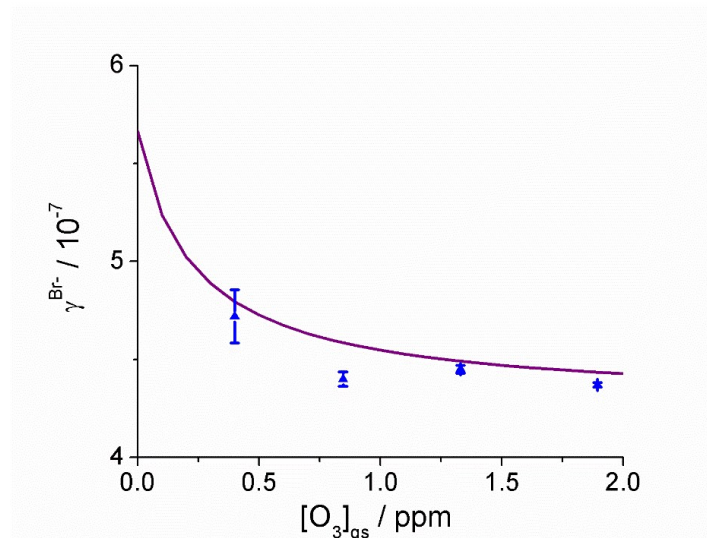## S.4.2 Study of the influence of pH on ozone by Br⁻ solutions and particles

Figure S3 shows the ozone uptake coefficients estimated by the "only Br⁻ model" for a 0.12 M NaBr solution, and the uptake coefficients obtained by Artiglia et al.[9] for a solution containing 0.12 M NaBr and 0.01 M HCl.

Figure S3. Ozone uptake coefficients estimated by the "only Br⁻ model" for a 0.12 M NaBr solution, and uptake coefficients obtained by Artiglia et al.[9] for a solution containing 0.12 M NaBr and 0.01 M HCl.

S.4.3 Two-Langmuir isotherm for O_3 adsorption

The number of O_3--H_2O sites (N_{O3}^{H2O}) for O_3 adsorption can be estimated as the difference $N_{O3,max}$ - [Y⁻]$_{surf}$, where $N_{O3,max}$ is the maximum number of sites for O_3 adsorption, and [Y⁻]$_{surf}$ is equal to the number of O_3--Y⁻ sites. According to literature data, $N_{O3,max}$ is estimated to be in the range $(0.24-6) \cdot 10^{14}$.[16, 17] Therefore, an intermediate value of $1 \cdot 10^{14}$ molecule cm⁻² will be assumed. As pointed out in the Model

Description section, a two-site Langmuir model can be used to describe adsorption of a molecule onto sites that have different adsorption energies. According to this model, the total concentration of adsorbed ozone ($[O_3]_{int,t}$) is given by the following equations:

$$[O_3]_{int,t} = [O_3]_{int,Y-} + [O_3]_{int,H_2O} = N_{O_3}^{Y-} \frac{K_{O_3}^{Y-}[O_3]_{gs}}{1 + K_{O_3}^{Y-}[O_3]_{gs}} + N_{O_3}^{H_2O} \frac{K_{O_3}^{H_2O}[O_3]_{gs}}{1 + K_{O_3}^{H_2O}[O_3]_{gs}}$$

(E23)

where $[O_3]_{int,Y-}$ is the concentration of O_3 adsorbed onto $Y^-_{(surf)}$, and $[O_3]_{int,H2O}$ is the concentration of O_3 adsorbed onto $H_2O_{(surf)}$. Parameter K_{O3}^{H2O}, whose definition can be found in the main text, has been determined to be $5 \cdot 10^{-16}$ cm^3 molecule^{-1} at ambient temperature.[18] Figure S4 shows the estimated values of $[O_3]_{int,H2O}$ and $[O_3]_{int,Br-}$ for different aqueous halide and $O_{3(gs)}$ concentrations, using equation E23 and the value of K_{O3}^{Y-} for 22 °C estimated with equation 16 from the main text.
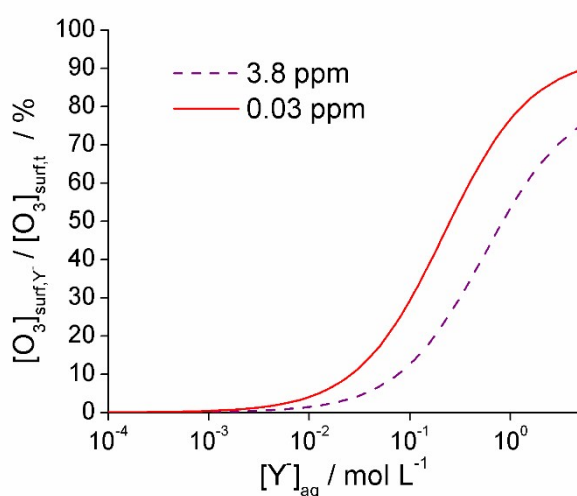

Figure S4. Estimated percentage of adsorbed O_3 that is adsorbed on Y^- (Y=I, Br, Cl) as a function of Y^- concentration at about 22 °C. As indicated in the text, adsorbed O_3 that is not adsorbed on Y^- is adsorbed on H_2O.

S.4.4 Results for conditions used in previous studies of the reactions Br⁻+O₃ using final models

Figure S5 shows final model results ("mixed Br⁻/Cl⁻ results") for the conditions used in previous studies of the interaction of O_3 with Br⁻ solutions or particles apart from results shown in the main manuscript. Experimental results are displayed also for comparison. In cases where no NaCl or HCl have been used, the "only Br⁻ model" and "mixed Br⁻/Cl⁻ model" yield the same results.

Other final model results not shown in the main manuscript are the following:

- An uptake coefficient of $2.0 \cdot 10^{-6}$ for 285 nm-sized 5.9 M NaBr aerosols exposed to 1.55 ppm O_3 at 25 °C (the experimental value is $2.3 \cdot 10^{-6}$).[19, 20]
- An uptake coefficient ratio of 3.6 for mixed 2 M NaCl + 0.03 M NaBr relative to a pure 0.03 M NaBr solution using $[O_3]_{gs}$=32 ppm at 17 °C (the experimental value is 3.2).[21]
- An uptake coefficient ratio of 2.5 for mixed 2 M NaCl + 0.12 M NaBr relative to a pure 0.12 M NaBr solution using $[O_3]_{gs}$=32 ppm at 17 °C (the experimental value is 2.7).[21]
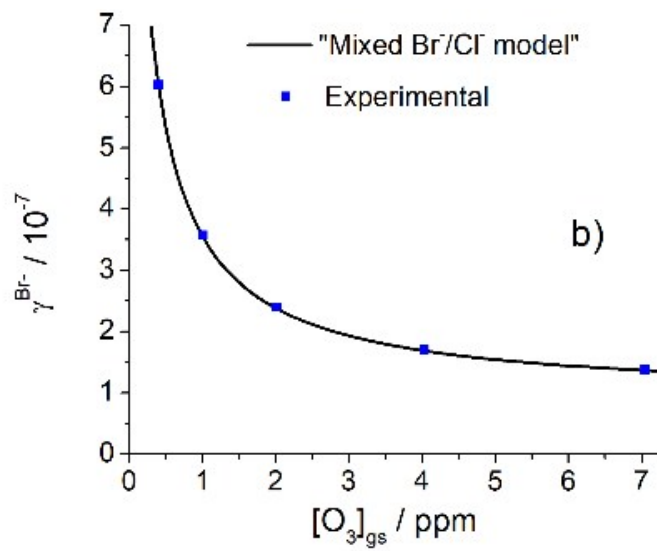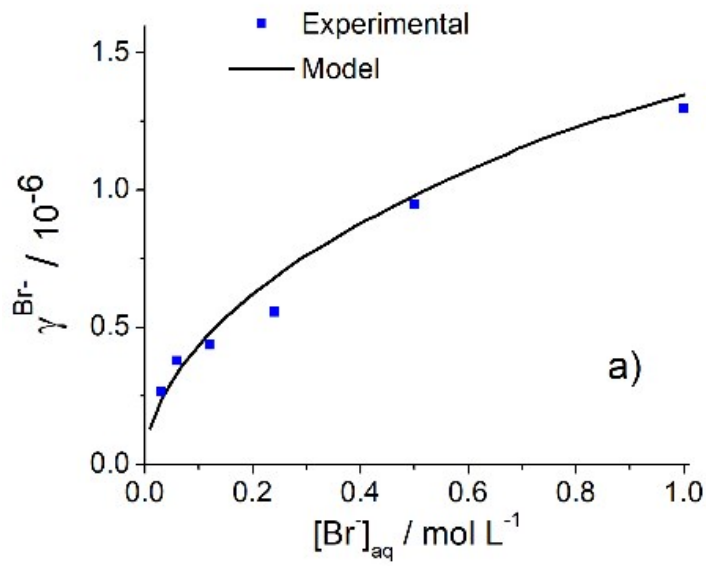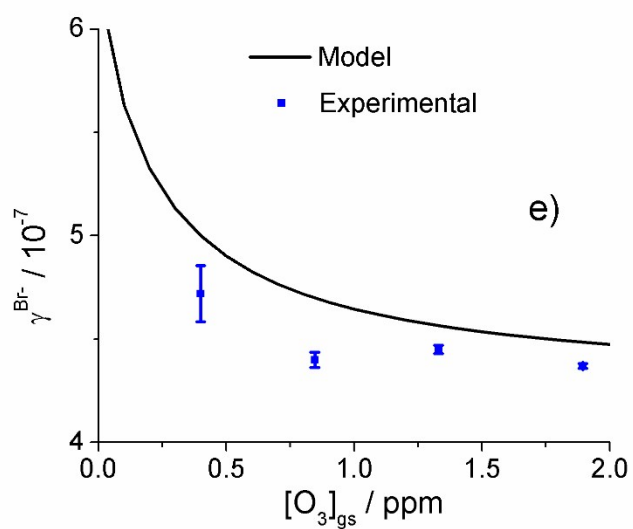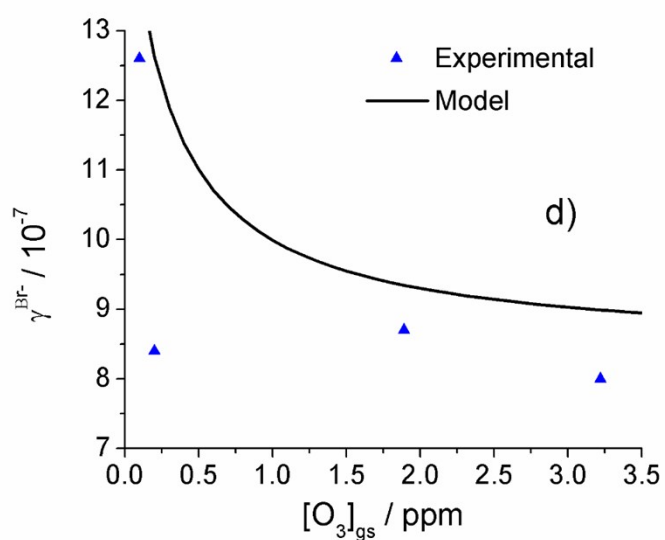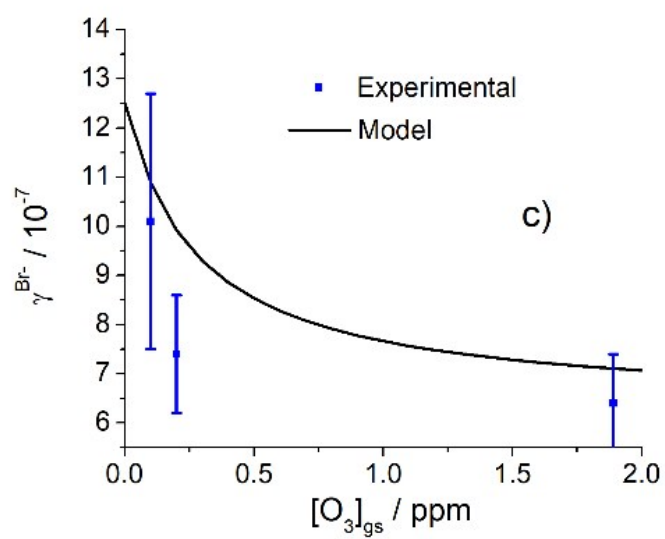
Figure S5. Comparison of experimental results of previous studies to results given by the "mixed Br⁻/Cl⁻ model" for the following conditions: a) pure NaBr solution, $[O_3]_{gs}$=1.8 ppm, 15.7 °C, pH 6 (compare

to Figure 3);[22] b) solution containing $8.5 \cdot 10^{-3}$ M NaBr, 0.55 M NaCl, and 0.01 M HCl, 0 °C, pH 2 (compare to Figure 4);[14] c) solution containing 0.12 M NaBr and 0.1 M HCl, 0 °C, pH 1 (compare to Figure 5);[9] d) solution containing 0.24 M NaBr and 0.1 M HCl, 1 °C, pH 1 (compare to figure 6);[9] and e) solution containing 0.12 M NaBr and 0.01 M HCl, 0 °C, pH 2 (compare to Figure S1).[9]

S.5 Interaction of O_3 with I^- and Cl^- in seawater

The ozone uptake coefficients estimated by the "mixed I^-/Cl^- model" for the O_3 and I^- concentrations used by Garland et al.[23] have been fit to the correlation expected for aqueous-phase reaction kinetics. This has been done, even though a surface reaction contribution is expected for these conditions, in order to draw an "apparent" rate constant of the aqueous-phase reaction and to compare this value with the one obtained by Garland et al.[23] As deduced from equation 8, the aqueous-phase reaction in solution obeys the following kinetics:

$$\frac{1}{\gamma_{aq}^{I^-}} = \frac{1}{\alpha_{aq}} + \frac{\omega}{4s\sqrt{k^{I^-} \cdot D_{aq}}} \frac{1}{a_{I^-}^{0.5}}$$

(E24)

Therefore, a correlation of $1/\gamma^{I^-}$ versus $1/a_{I^-}^{0.5}$ should yield a line of slope $\omega/(4sk_{I^-}^{0.5}D_{aq}^{0.5})$ if the aqueous-phase reaction dominates uptake and if there is no contribution from the surface reaction, and the value of the slope can be used to obtain k^{I^-}. The model-estimated values of these variables are plotted in Figure S6 ("mixed I^-/Cl^-" model) for the concentrations used by Garland et al.[23] The linear correlation has a regression coefficient of 0.98 and its slope yields an apparent rate constant of $3.1 \cdot 10^9$ M^{-1} s^{-1}. When $[Cl^-]_{surf}$ is decreased by a factor of 2.1, the resulting rate constant matches the value obtained by Garland et al.[23] ($2 \cdot 10^9$ L mol^{-1} s^{-1}) and the line has a slope of 0.999. Therefore, this value of $[Cl^-]_{surf}$ has been assumed to model surface reaction in seawater.
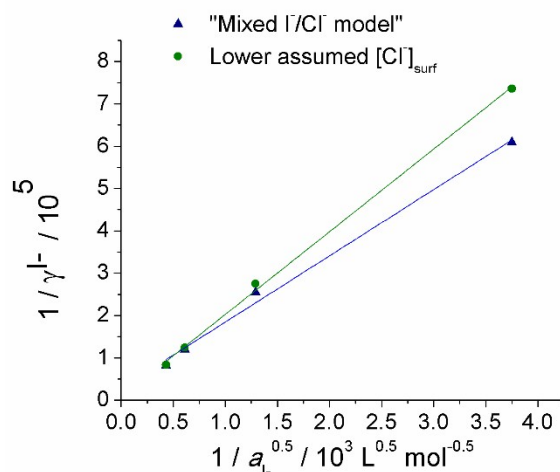

Figure S6. Linear fit of model-estimated uptake coefficients for the I^- and O_3 concentrations (0.1 ppm) used by Garland et al.[23] to the correlation expected for aqueous-phase kinetics employing two different assumptions about the ratio $[Cl^-]_{surf}/[Cl^-]_{aq}$: it is equal to the value estimated in the main manuscript ("mixed I^-/Cl^- model") or it is 2.1 times lower ("lower assumed $[Cl^-]_{surf}$").

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