

# IUPAC Task Group on Atmospheric Chemical Kinetic Data Evaluation – Data Sheet VI.A4.24 HET\_SL\_24

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## ClONO<sub>2</sub> + HCl (aqueous sulphuric acid aerosol) → HONO<sub>2</sub> + Cl<sub>2</sub>

### Experimental data

Parameter	[H <sub>2</sub> SO <sub>4</sub> ] (wt%)	[HCl] / [HNO <sub>3</sub> ] M	Temp./K	Reference	Technique/ Comments
$\gamma(\text{ClONO}_2)$					
3 x 10 <sup>-3</sup>	65%		210	Tolbert, Rossi and Golden, 1988	Knud-MS (a)
2 x 10 <sup>-3</sup>	40		218	Hanson and Ravishankara, 1991	WWFT-CIMS (b)
2 x 10 <sup>-4</sup>	60		215		
<1 x 10 <sup>-4</sup>	65		215		
<2 x 10 <sup>-5</sup>	70		220		
<2 x 10 <sup>-5</sup>	75		230		
0.038	46.6	< 10 <sup>-4</sup>	202	Hanson and Ravishankara, 1994	WWFT-CIMS (c)
0.10	46.6	10 <sup>-3</sup>			
0.30	46.6	6 x 10 <sup>-3</sup>			
0.16±0.06	49	5.8 x 10 <sup>-3</sup>	197	Zhang, Leu and Keyser, 1994	WWFT-CIMS (d)
0.10±0.05	51	2.2 x 10 <sup>-3</sup>	198.5		
0.04±0.013	55	3.3 x 10 <sup>-4</sup>	201		
(9.5±3.2) x 10 <sup>-3</sup>	60	2.5 x 10 <sup>-5</sup>	205.5		
(1.8±1.6) x 10 <sup>-3</sup>	65	2.5 x 10 <sup>-6</sup>	211		
0.3	43	8.2 x 10 <sup>-4</sup>	194	Elrod et al., 1995	WWFT-MS(e)
0.083	47	1.4 x 10 <sup>-4</sup>	196		
0.038	51	2.4 x 10 <sup>-5</sup>	198		
0.01	55.2	3 x 10 <sup>-6</sup>	201		
0.11	20.2	4.88 x 10 <sup>-4</sup> (28.35% HNO <sub>3</sub> )	195		
0.058	44		197		
0.021	50.3	6.18 x 10 <sup>-5</sup> (6.1% HNO <sub>3</sub> )	199		
		1.38 x 10 <sup>-5</sup> (2.26% HNO <sub>3</sub> )			
0.07	45	10 <sup>-8</sup> mbar	203-205	Hanson, 1998	WWFT-CIMS (f)
0.6	45	10 <sup>-6</sup> mbar			
0.03	51	10 <sup>-8</sup> mbar			
0.019	51	10 <sup>-6</sup> mbar			
0.011	55	10 <sup>-8</sup> mbar			
0.043	55	10 <sup>-6</sup> mbar			
0.18±0.10	44	10 <sup>-6</sup> mbar, 4.4% HNO <sub>3</sub>	205		
	20.3				
0.25±0.15		10 <sup>-6</sup> mbar, 25.6% HNO <sub>3</sub>			
0.38±0.1	49±1	1.1 x 10 <sup>-7</sup> mbar	240	Hanson, 1998	AFT-CIMS(g)

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### Comments

- (a) The H<sub>2</sub>O vapor pressure in the flow reactor was 5.3x10<sup>-4</sup> mbar (±30%). Before ClONO<sub>2</sub> uptake at typical pressures of 6.4x10<sup>-3</sup> mbar, the 65% H<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O solution was exposed to 1.7x10<sup>-3</sup> mbar HCl for an hour to result in an estimated mole fraction ratio HCl:H<sub>2</sub>O=2x10<sup>-4</sup>. More than 85% of the HCl is recoverable as Cl<sub>2</sub> from the 65% H<sub>2</sub>SO<sub>4</sub> solution at 210K. Resulting  $\gamma$  values are very similar to experiments without added HCl.
- (b) The cold aqueous solution of H<sub>2</sub>SO<sub>4</sub> was flowed down the walls of the vertical flow tube with a liquid film residence time of 20-30s. p(H<sub>2</sub>O) was generally around 1.3 x 10<sup>-3</sup> mbar, [HCl] for the experiments on the 40% solution was 2 x 10<sup>11</sup>, for the 60 to 75% solution was in the range 1 to 3 x 10<sup>11</sup> molecule cm<sup>-3</sup>. The error limits are reported to be (+100%,-50%) except for the 40 and the 60% solution. The uptake of ClONO<sub>2</sub> in the presence of HCl is not accelerated compared to experiments without HCl. A number of experiments were performed at high HCl concentrations with the result that  $\gamma$  is significantly enhanced at comparable ClONO<sub>2</sub> concentrations: for a 60% solution  $\gamma$  was 2 x 10<sup>-4</sup> (215K), 0.006 (215K) and ~0.01 (238K) for gas phase HCl concentrations of 2 x 10<sup>10</sup>, 7 x 10<sup>12</sup> and ~5 x 10<sup>12</sup> molecule cm<sup>-3</sup>.
- (c) Liquid H<sub>2</sub>SO<sub>4</sub> (46.6% to 65%) was applied as a cold liquid to the inner wall of the flow tube. The HCl was either added to the liquid (46.6 and 51%) or taken up from the gas phase (55.6, 57.5, 58.5, 59.8 and 65%). No significant temperature dependence of the uptake coefficient in the temperature range 192 to 208 K was observed. The uptake coefficient increased with increasing p(HCl) and increasing H<sub>2</sub>O activity. Significant uptake was observed at [HCl] → 0 which was attributed to the ClONO<sub>2</sub> hydrolysis reaction. It was found from fitting experimental data over a wide range that a surface-mediated reaction between Cl<sup>-</sup> and ClONO<sub>2</sub> scaling with p(HCl) was required in addition to the competitive bulk reaction to account for the observed uptake rates.
- (d) Total pressure of He 0.67 mbar and flow velocity 890 cm s<sup>-1</sup> linear. The partial pressure of H<sub>2</sub>O (5ppm at 100mbar) was held constant throughout the range of 195 to 220K in order to control [H<sub>2</sub>SO<sub>4</sub>] between 45 and 70% as a function of temperature. p(ClONO<sub>2</sub>) and p(HCl) were varied between (1 - 2.5) x 10<sup>-7</sup> and (2.7 - 27) x 10<sup>-7</sup> mbar, respectively. The value of  $\gamma$  increases by a factor of ten with increasing [HCl] in the given range. Identical values of  $\gamma$  were measured on liquid sulfuric acid containing up to 5% of HNO<sub>3</sub> and 5 ppm. H<sub>2</sub>O
- (e) Reactive uptake experiment using a vertically-mounted wetted-wall flow reactor coupled to a differentially-pumped beam-sampling quadrupole MS. The substrates were bulk ternary or quaternary solutions of H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O-HCl-HNO<sub>3</sub>. The uptake measurements were performed at 203 K, and the composition of the solutions selected to correspond to various atmospheric equilibrium temperatures in the range 194-201K. p(HCl) over the solutions was measured at temperatures in the range 208 - 233K to determine H\*(HCl) and p(ClONO<sub>2</sub>) was of the order of 1.3 x 10<sup>-6</sup> mbar. It was established that  $\gamma_{ss}$  was essentially independent of temperature at a given composition and that it was controlled by the HCl solubility in the condensed phase.
- (f) Uptake experiment in rotating wetted wall flow tube using CIMS detection for ClONO<sub>2</sub>, Cl<sub>2</sub>, HOCl and water vapor. HCl was doped into the 49.5, 51, 53 and 55% H<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O solutions from the gas phase. For 45% sulfuric acid solutions HCl was added by mixing pure with sulfuric acid containing known amounts of HCl. The measured uptake is the sum of the reactions of ClONO<sub>2</sub> with HCl and the hydrolysis with H<sub>2</sub>O. The parametrization of the total

uptake contains an interfacial term for  $\text{ClONO}_2 + \text{HCl}$  which is important for  $[\text{HCl}] > 10^{-3}$  M. For liquid ternary solutions containing  $\text{HNO}_3$  the uptake was significantly lower compared to pure sulfuric acid solutions: for partial pressures corresponding to 5 ppm  $\text{H}_2\text{O}$  and 5 ppb  $\text{HNO}_3$  at 19 km altitude  $\gamma_{\text{ss}}$  was typically lower by a factor of two.

- (g) Uptake on sub-micron aerosol particles of  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  (49±1 wt %) doped with HCl, measured at 240K and 120 mbar total pressure to determine surface accommodation coefficient,  $\alpha_s$ . The surface-area-weighted mean diameter was 160 nm and the number density (2 - 5)  $\times 10^5$   $\text{cm}^{-3}$ . The particles were doped with HCl ( $p(\text{HCl}) = 1.1$  and  $2.0 \times 10^4$  mbar), resulting in an HCl content of  $9.6 \times 10^{-3}$  and  $1.7 \times 10^{-2}$  M, respectively. The weak dependence of  $\gamma$  on  $p(\text{HCl})$  suggests that reaction rate is not quite fast enough to force the measured  $\gamma$  to the value of  $\alpha_s$ . A best estimate for  $\alpha_s$  based on a crude analysis gave  $0.72 \pm 0.1$  but a value of  $\alpha_s = 1$  is consistent with the measurements.

### Preferred Values

Parameter	Value	T/K
$\alpha_s$	1.0	298
$b_0 / \text{M}^2 \text{cm}^{-2}$	$7.9 \times 10^{-11}$	
$k_{\text{H}_2\text{O}} / \text{s}^{-1}$	$1.95 \times 10^{10} \exp(2800/T)$	190 - 280
$k_{\text{H}^+} / \text{M}^{-1} \text{s}^{-1}$	$1.22 \times 10^{12} \exp(6200/T)$	190 - 280
$\Gamma_s$	$66.12 \exp(-1374/T) \times H_{\text{ClONO}_2} \times M_{\text{HCl}}$	
$H_{\text{ClONO}_2} / \text{M} \cdot \text{bar}^{-1}$	$1.6 \times 10^{-6} \exp(4710/T) \exp[(-0.306 + 24.0/T) M_{\text{H}_2\text{SO}_4}]$	200 - 280
$D_l / \text{cm}^2 \text{s}^{-1}$	$5 \times 10^{-8} T / \eta$	180 - 240
.		
<i>Reliability</i>		
$\Delta \log(\alpha)$	$\pm 0.1$	298
$\Delta \log(\gamma)$	$\pm 0.3$	200 - 280

$M_{\text{H}_2\text{SO}_4}$  = molarity  $\text{H}_2\text{SO}_4$  ( $\text{Mol dm}^{-3}$ );  $M_{\text{HCl}}$  = molarity HCl ( $\text{Mol dm}^{-3}$ ) =  $p(\text{HCl}) \times H_{\text{HCl}}^*$ .

### Comments on Preferred Values

There is a large body of experimental data on the reactive uptake of  $\text{ClONO}_2$  in the presence of HCl on  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  surfaces, mainly covering the range of temperature and humidity, and acid composition relevant for the upper troposphere and lower stratosphere. The uptake leads to efficient formation of  $\text{HNO}_3$  and  $\text{Cl}_2$  products both of which transfer rapidly to the gas phase. At low  $p(\text{HCl})$ , when hydrolysis of  $\text{ClONO}_2$  contributes,  $\text{HOCl}$  is also observed as a product.

The kinetic results are generally consistent between the different studies which used both bulk and dispersed (aerosol) surfaces, although the uptake coefficients measured on bulk surfaces often required large correction for gas phase diffusion effects, because of fast reactive uptake of  $\text{ClONO}_2$  (or HCl) at the surface. The reactive uptake coefficient uptake increases strongly with  $\text{H}_2\text{O}$  content of  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  solutions in the corresponding range 40 - 70 wt% at constant  $p(\text{HCl})$ , which reflects the increasing solubility of HCl in  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  as relative humidity increases. The uptake coefficient shows only a very weak dependence on temperature.

The uptake coefficient also shows a complex dependence on  $p(\text{HCl})$ , or on  $[\text{HCl}]_l$  in experiments where the solutions were specifically prepared. At intermediate  $[\text{HCl}]$  Elrod et al

(1997) found  $\gamma$  proportional to  $[\text{HCl}]^{1/2}$ , whilst Zhang et al (1994) who used higher  $p(\text{HCl})$ , gave close to linear dependence on  $[\text{HCl}]$ . Hanson and Ravishankara (1994) covered a wider range of  $p(\text{HCl})$ , and found that for intermediate and high HCl amounts,  $\gamma$  values were proportional to  $p(\text{HCl})^{1/2}$  and  $p(\text{HCl})$  respectively, attaining a  $\gamma$  value of 0.3 at  $1.3 \times 10^{-4}$  mbar HCl at 202 K. At low  $p(\text{HCl})$ ,  $\gamma$  tends to a constant value of  $\sim 0.01$ , which is due to the additional contribution of hydrolysis of  $\text{ClONO}_2$  to the overall  $\gamma$ . The observed effects of temperature can also be rationalised by its effect on HCl solubility in  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  solution of varying mole fraction, together with compensating effects on the rate coefficient of the liquid phase reaction. At low temperatures the presence of  $\text{HNO}_3$  in the  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  solutions leads to significantly lower  $\gamma$  relative to solutions without  $\text{HNO}_3$  (Hanson,1998). The effect increases with  $[\text{HNO}_3]/[\text{H}_2\text{SO}_4]$  and at equal wt% amounts to a 50% reduction in  $\gamma$  at 205 K.

Hanson and Ravishankara (1994) suggested that two pathways control the rate of heterogenous reaction of  $\text{ClONO}_2$  with HCl: a direct surface reaction; and a bulk phase reaction involving reaction of solvated  $\text{ClONO}_2$  with  $\text{Cl}^-$ , leading to the distinct kinetic dependencies on  $p(\text{HCl})$ . Hanson (1998) has reanalysed their earlier data, and those of Elrod et al.(1997) and Zhang et al.(1994) in the light of improved measurements of HCl solubility, and has formulated a parameterisation for overall  $\gamma$  taking into account the different reactive processes, using a resistance-model formulation, eq (1):

$$\frac{1}{\gamma} = \frac{1}{\alpha_s} + \frac{1}{\frac{1}{\frac{1}{\Gamma_b} + \frac{1}{\alpha_s \frac{k_{sol}}{k_{des}}}} + \Gamma_s} \quad (1)$$

Hanson's parameterisation gives overall  $\gamma$  for uptake of chlorine nitrate as a function of mole fraction of  $\text{H}_2\text{SO}_4$  and  $p(\text{HCl})$  for stratospheric conditions near 200 K, using an empirical representation of the composition-dependent liquid phase resistance due to chemical reaction.

Shi et al. (2001) have adopted the Hanson model and extended the parameterisation to include temperature dependence, using re-evaluated  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  composition and temperature dependent expressions for HCl solubility,  $H^*_{\text{HCl}}$  (see VI.A4.14), chlorine nitrate solubility,  $H_{\text{ClONO}_2}$ , and resistance term  $\Gamma_b$  for the  $\text{ClONO}_2 + \text{H}_2\text{O}$  reaction (see VI.A4.25). Shi et al. (2001) assumed that the  $\text{ClONO}_2 + \text{HCl}$  reaction is  $\text{H}^+$  catalysed and its rate is diffusion controlled, as deduced by Hanson (1998). They also assume that the direct surface reaction of  $\text{ClONO}_2$  with HCl depends on bulk-liquid  $[\text{ClONO}_2]$  and  $[\text{HCl}]$  through their respective  $T$ - and composition-dependent  $H$  values. Overall the deviation from the experimental data was 40% for the Shi model compared to 46% for the Hanson (1998) parameters. We adopt the Shi et al. (2001) parameters in this recommendation.

The preferred value  $\alpha_s$  is based on the measured uptake coefficients on HCl-doped aerosols reported by Hanson (1998), which support a surface accommodation coefficient near unity ( $\alpha_s = 1$ . see comment *g* above). It follows that the term  $\alpha_s k_{sol}/k_{des}$  which is equivalent to the surface to bulk resistance term,  $\Gamma_{sb}$ , must also be close to unity, and equation (1) reduces to:

$$\frac{1}{\gamma} = 1 + \frac{1}{(\Gamma_b + \Gamma_s)} \quad (2)$$

The resistance term  $\Gamma_b$  for the bulk reaction probability is given by:  $\Gamma_b = 4RTH(D_1k^I)^{1/2}/c$  where  $D_1$  and  $H$  are respectively the diffusion coefficient and Henry's constant for ClONO<sub>2</sub>.  $k^I$  is the sum of the first order rate coefficients for reaction of ClONO<sub>2</sub> with Cl<sup>-</sup> ( $k_{HCl}^I$ ) and H<sub>2</sub>O ( $k_{H_2O}^I$ ) in the bulk. Combining the resistance terms for these two processes acting in parallel gives:

$$\Gamma_b = \Gamma_{H_2O} \sqrt{1 + \frac{k_{HCl}^I}{k_{H_2O}^I}} \quad (3)$$

Here  $\Gamma_{H_2O} = 4RTH(D_1k_{H_2O}^I)^{1/2}/c$  and the recommended temperature and composition dependence of the first order rate coefficient,  $k_{H_2O}^I$ , and solubility of ClONO<sub>2</sub>,  $H$ , are given in the functional forms described in the data sheet for the ClONO<sub>2</sub> + H<sub>2</sub>O reaction (see VI.A4.25). The overall hydrolysis rate constant is given by the expression:

$$k_{H_2O}^I (s^{-1}) = (k_{H_2O} + k_{H^+} a_{H^+}) a_w$$

where  $a_w$  = activity water;  $a_{H^+}$  = acid activity (Mol dm<sup>-3</sup>).  $a_w$  is calculated from:  $a_w = \exp[(-69.775X - 18253.7X^2 + 31072.2X^3 - 25668.8X^4)/(1/T - 26.9033/T^2)]$  where wt = weight% H<sub>2</sub>SO<sub>4</sub>.

The diffusion controlled rate coefficient for the ClONO<sub>2</sub> + HCl reaction is given by the expression:

$$k_{HCl}^I (s^{-1}) = b_0 D_1 a_{H^+} H_{HCl}^* p(HCl) \quad (4)$$

The recommended value for the constant  $b_0$  ( $= 7.9 \times 10^{-11} \text{ M}^2\text{cm}^2$ ), was determined from the experimental measurement of the reacto-diffusive length at 250 K at high [HCl] (Hanson 1998). The value of  $H_{HCl}^*$  is given as a function of H<sub>2</sub>SO<sub>4</sub> mole fraction,  $X$ , by the expression:

$$H_{HCl}^* = (0.094 - 0.61X + 1.2X^2) \exp(-8.68 + (8515 - 10718X^{0.7})/T) \quad (5)$$

as given in data sheet for HCl solubility VI.A4.14. The acid activity is derived from the data from the thermodynamic model of Carslaw et al.(1997). For the expression for  $a_{H^+}$  in the table the Carslaw model was extended to include the acidity of pure water, to provide values of  $a_{H^+}$  as a function of acid wt% (wt) in units of M dm<sup>-3</sup> extending to dilute solution (wt ~ 0%):

$$a_{H^+} = \exp[60.51 - 0.095\text{wt} + 0.0077\text{wt}^2 - 1.61 \times 10^{-5}\text{wt}^3 - (1.76 + 2.52 \times 10^{-4}\text{wt}^2)T^{0.5} + (-805.89 + 2.53.05\text{wt}^{0.076})/T^{0.5}]$$

Diffusion coefficients,  $D_i$ , are calculated using the expression:  $D_i = CT/\eta$ , with  $C = 5 \times 10^{-8} \text{ cm}^2\text{cP K}^{-1}\text{s}^{-1}$  (taken from Klassen et al., 1998). Viscosity data for H<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O obtained by Williams and Long(1995) was reparameterized to give a more general formulation for  $D_i$  covering the temperature range 200 – 300 K and 0 – 80wt% H<sub>2</sub>SO<sub>4</sub>:

$$\eta = AT^{1.43} \exp(448/T - T_0)$$

$$A = 169.5 + 5.18\text{wt} - 0.0825\text{wt}^2 + 3.27 \times 10^{-3}\text{wt}^3$$

$$T_0 = 144.11 - 0.166\text{wt} + 0.015\text{wt}^2 - 2.18 \times 10^{-4}\text{wt}^3$$

The resistance term  $\Gamma_s$  for the surface reaction probability was parameterised by Shi et al (2001) as follows:

$$\Gamma_s = 66.12 \exp(-1374/T) H_{\text{ClONO}_2} H_{\text{HCl}}^* P_{\text{HCl}} \quad (6)$$

which assumes a dependence of surface reaction rate on liquid phase concentrations of reactants, which is determined by their solubilities. An Arrhenius temperature dependence is assumed and the recommended constant term was evaluated by fitting to the ClONO<sub>2</sub> uptake rate data at high [HCl]. This expression gave a better representation of the temperature dependence of the surface reaction component than that used by Hanson (1998).

The figure shows a comparison of  $\gamma$  values calculated using eq (2) for a large range of p(HCl) and at different acid wt% H<sub>2</sub>SO<sub>4</sub>, with experimental data obtained mainly at temperatures near 200 K. The model gives a good fit to the data over the range of conditions employed and will give reasonably accurate values of  $\gamma(\text{ClONO}_2)$  for UTLS conditions. Overall the deviation reported by Shi et al. (2001) from experimental data used for the fit was 40%, compared with 46% for the parameters reported by Hanson (1998). These form the basis of the recommended uncertainty in  $\gamma$  over the temperature range 200 – 280 K. The temperature dependence of the  $\gamma_r$  is in fact rather small due to compensating effects of the temperature dependence of  $D_1$  and  $H_{\text{HCl}}^*$ . For detailed discussion of the model/measurement comparison the Shi et al paper (2001) should be consulted.

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**Figure 1:** Plot of  $p(\text{HCl})$  dependence of uptake coefficient at different  $\text{H}_2\text{SO}_4$  wt%. Symbols show experimental data: open circles = 45-47%; closed circles = 49-49.5%; open squares = 51%; triangles = 55%; half filled squares = 60%; square cross = 65%. Lines show predicted uptake coefficients using recommended parameters for each wt%. Temperature = 197 - 205 K, except data collected for  $p(\text{HCl}) = \sim 10^{-7}$  bar on aerosols by Hanson (1998) at 240 K

