

IUPAC Task Group on Atmospheric Chemical Kinetic Data Evaluation – Data Sheet V.A2.8 MD8

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HNO₃ + mineral oxide (dust) surfaces

Experimental data

<i>Parameter</i>	Temp./K	Reference	Technique/ Comments
<i>Uptake coefficients: γ, γ_0</i>			
$\gamma_0 = 0.09$ (CaCO ₃ , dried)	298	Fenter et al., 1995	Knudsen-MS (a)
$\gamma_0 = 0.15 \pm 0.03$ (CaCO ₃ , non-dried)			
$\gamma_0 = 1.4 \times 10^{-5}$ (CaCO ₃ , dried)	298	Underwood et al., 2000	Knudsen-MS DRIFTS (b)
$\gamma_0 = (2.5 \pm 0.1) \times 10^{-4}$ (CaCO ₃ , dried)	295	Goodman et al, 2000	Knudsen-MS DRIFTS (c)
$\gamma_0 = 0.10 \pm 0.025$ (CaCO ₃ , heated)	298	Hanisch and Crowley, 2001a	Knudsen-MS (d)
$\gamma_0 = 0.18 \pm 0.045$ (CaCO ₃ , non-heated)			
$\gamma_0 = 0.13 \pm 0.033$ (Al ₂ O ₃)			
$\gamma_0 = 0.11 \pm 0.03$ (Saharan dust)			
$\gamma_0 = 0.06 \pm 0.015$ (Arizona dust)			
$\gamma_0 = 0.14$ (Saharan dust)	296	Hanisch and Crowley, 2001b	Knudsen-MS (e)
$\gamma_0 = 0.17$ (Chinese dust)			
$\gamma_0 = (9.7 \pm 0.5) \times 10^{-5}$ (α -Al ₂ O ₃)	298, 295	Underwood et al., 2001a,b	Knudsen-MS (f)
$\gamma_0 = 5.2 \times 10^{-5}$ (Gobi dust)			
$\gamma_0 = (2.0 \pm 0.1) \times 10^{-5}$ (Saharan sand)			
$\gamma_0 = (5.3 \pm 0.3) \times 10^{-5}$ (α -Fe ₂ O ₃)			
$\gamma_0 = (2.9 \pm 0.2) \times 10^{-5}$ (SiO ₂)			
$\gamma = (1.6 \pm 0.3) \times 10^{-9}$ (SiO ₂)	296	Goodman et al., 2001	FTIR (g)
$\gamma = (4 \pm 1) \times 10^{-8}$ (α -Al ₂ O ₃)			
$\gamma_0 = (1.5 \pm 1.0) \times 10^{-5}$ (γ -Fe ₂ O ₃)	297	Frinak et al., 2004	Knudsen-MS/FTIR (h)
$\gamma_0 = 2.9 \times 10^{-5}$ (γ -Fe ₂ O ₃)	220		
$\gamma_0 = 0.13 \pm 0.02$ (α -Al ₂ O ₃)	298	Seisel et al., 2004	Knudsen- MS/DRIFTS (i)
$\gamma_0 = 0.11 \pm 0.02$ (Sahara)			
$\gamma_0 = 2 \times 10^{-3}$ (CaCO ₃ , dried)	296	Johnson et al, 2005	Knudsen-MS (j)
$\gamma = 0.11$ (Arizona dust, RH 33 %)		Vlasenko et al., 2006	AFT (k)
$\gamma = 0.11$ (CaCO ₃ , RH 33 %)			
$\gamma < 5 \times 10^{-4}$ (SiO ₂ , RH 33 %)			
$\gamma_0 = 0.01$ (CaCO ₃ , polished marble)	300	Santschi and Rossi, 2006	Knudsen-MS (l)
$\gamma_0 = 0.04$ (CaCO ₃ , cut marble)			
$\gamma_0 = 0.3$ (CaCO ₃ , powder)			
$\gamma_0 > 8 \times 10^{-5}$ (Na-montmorillonite, RH 29%)	210-232	Mashburn et al., 2006	Knudsen-MS /FTIR (m)
$\gamma_0 = 4 \times 10^{-4}$ (Na-montmorillonite, RH 44%)			
$\gamma \geq 0.06$ (40 % RH)	298	Liu et al., 2008	(n)
$\gamma = 0.21$ (80 % RH)			
<i>Accommodation coefficient: α_s</i>			
$\alpha_s = 1.0$ (Arizona dust, RH 33 %)	298	Vlasenko et al., 2009	AFT (o)

Comments

- (a) Irreversible uptake observed with H₂O and CO₂ detected as gas-phase products of reaction of HNO₃ with bulk samples. Comparison of results using powder and pressed pellet samples revealed no dependence of the uptake coefficient on the internal surface area, hence the geometric surface area was used to derive the value listed in the Table. Under the experimental conditions employed ([HNO₃] = 10¹⁰-10¹³ molecule cm⁻³) the uptake coefficient decreased slowly with time of exposure, and initial uptake coefficients (γ_0) are reported. Pellet samples that had been dried (under vacuum) for several hours were less reactive than non-dried samples.
- (b) The uptake coefficients were derived from experiments with [HNO₃] \approx 1x10¹¹ molecule cm⁻³. Linear dependence of uptake coefficient on mass observed, and taken to justify use of the BET surface area to calculate the uptake coefficient. An uptake coefficient of 1 x 10⁻³ was derived when the geometric surface area was assumed.
- (c) Bulk dust samples were prepared by drying a water slurry. The uptake coefficients were derived from experiments with [HNO₃] \approx 1x10¹² molecule cm⁻³ using a pore diffusion calculation for account for uptake to internal surfaces of the bulk samples. Heating and evacuating the samples overnight resulted in significantly lower values of γ .
- (d) Bulk dust samples were prepared by mixing the powder sample to a paste with ethanol or H₂O, and were usually treated by evacuation for 5 hours at 363 K. Four different grain sizes of Al₂O₃ were used, all yielding the same initial uptake coefficient. The data were thus analysed using the geometric surface area, and ignoring the contribution of internal surfaces. The initial HNO₃ concentration was varied between 6.5 x 10¹⁰ and 1.5 x 10¹² molecule cm⁻³. For Saharan dust, the same value of γ was obtained whether the sample was heated or not.
- (e) Same methods as (d) but better sensitivity enabled and lower HNO₃ concentrations to be used (10⁹ molecule cm⁻³). In contrast to (c), most of the samples were not heated after dispersion. In addition to those listed, initial uptake coefficients were also obtained for a series of clay minerals: Kaolinite (0.11), ripidolite (0.10), illite (0.11), illite/smectite (0.09), Ca-montmorillonite (0.11), Ca-montmorillonite (0.08), palygorskite (0.20), dolomite (0.14) and orthoclase (0.08). Three different grain sizes of Chinese dust were used, all yielding the same initial uptake coefficient, as did use of different sample depths. The data were thus analysed using the geometric surface area, and ignoring the contribution of internal surfaces. For Chinese dust, the same value of γ was obtained whether the sample was heated or not.
- (f) See (c). Bulk dust samples were prepared by spraying an aqueous slurry onto the heated sample holder and kept under vacuum overnight prior to an experiment. Experiments were carried out in the linear mass dependent regime, so that γ was calculated using the BET surface area of the dust sample. The effect of heating the sample was found to reduce the uptake coefficient by a factor of 10 for Gobi dust. A strong increase in the uptake coefficient with decreasing nitric acid was observed for α -Al₂O₃. A kinetic model of surface saturation and diffusion through the surface layers showed that the uptake coefficient obtained this way may be underestimated by factors of between 5 and 60, depending on e.g. the initial HNO₃.
- (g) Dry samples. The uptake coefficients were derived from time dependence of surface nitrate formation at high HNO₃ (10¹⁴ molecule cm⁻³). The rate of HNO₃ uptake to α -Al₂O₃ and CaO increased nearly 50-fold going from 0 % to 20 % humidity. The authors suggest that the results obtained are lower limits to γ due e.g. to the use of high HNO₃. A value of $\gamma = (4 \pm 1) \times 10^{-7}$ was obtained for CaO.
- (h) Bulk dust samples were prepared from an aqueous (or CH₃OH) slurry and dried under vacuum. Initial [HNO₃] \approx 3 x 10¹¹ molecule cm⁻³. Specific surface area of sample (measured using H₂O adsorption) used to calculate the uptake coefficient. Surface nitrate was observed following exposure of γ -Fe₂O₃ to HNO₃.

- (i) Bulk dust samples were prepared from an aqueous slurry and dried at room temperature under vacuum. $[\text{HNO}_3]$ was varied between 4×10^{11} and 1×10^{13} molecule cm^{-3} . The value of γ listed for Saharan dust is that obtained at low $[\text{HNO}_3]$. No dependence of γ on the mass of the substrate was observed, confirming that the geometric surface area was appropriate for calculating the uptake coefficient. For $\alpha\text{-Al}_2\text{O}_3$ the uptake coefficient decreased as $[\text{HNO}_3]$ was increased, and H_2O was released into the gas phase with a yield of unity.
- (j) Experiments used multiple, single or fractional layers of the sample. Initial $[\text{HNO}_3]$ was 6.5×10^{10} molecule cm^{-3} . The value of γ_0 is larger (factor of 10) than previously reported by the same group (Goodman et al., 2000) due to reduction of saturation effects. Results were also obtained for dolomite for which $\gamma = (6 \pm 4) \times 10^{-4}$ (average of multi-, single- and fractional layer experiments) was obtained. Geometric uptake coefficients for CaCO_3 were observed to show a dependence on sample mass, maximising at values of ≈ 0.03 .
- (k) HNO_3 (initial concentration $\approx 10^{11}\text{-}10^{12}$ molecule cm^{-3}) detected using radioactive labelling (H^{13}NO_3). Sub-micron Arizona dust aerosol was introduced into reactor via dry dispersion from its powder, CaCO_3 aerosol was generated by nebulising a saturated aqueous solution of CaCO_3 and diffusion drying. Particle number and size distribution was analysed using SMPS. The uptake coefficient was calculated using the time- and aerosol area dependent change in aerosol phase ^{13}N . Uptake coefficient on Arizona dust was observed so show a dependence on relative humidity ($\gamma = 0.02$ at $\text{RH} = 12\%$, increasing to $\gamma = 0.11$ at 73% RH when $[\text{HNO}_3]$ was 10^{12} molecule cm^{-3}).
- (l) Samples were cut and sometimes polished marble disks or CaCO_3 powder. H_2O (but not CO_2) observed as reaction product. $[\text{HNO}_3]_0 \approx (3\text{--}7) \times 10^{11}$ molecule cm^{-3} . Partial HNO_3 desorption from marble surfaces was observed, suggesting that part of the initial uptake was due to physisorption.
- (m) Na-montmorillonite samples exposed to $[\text{HNO}_3] > 3 \times 10^{12}$ molecule cm^{-3} . Uptake coefficients were extracted from IR surface analysis of nitrate formation (providing lower limits) and MS analysis of the gas-phase. In both cases the BET surface area was used. A strong dependence ($>$ factor of ten) of uptake coefficient on the relative humidity was observed.
- (n) Suspended CaCO_3 particles ($\approx 0.8 \mu\text{m}$) exposed to HNO_3 ($\approx 2\text{-}6 \times 10^{11}$ molecule cm^{-3}) in humidified air. Uptake coefficient derived from rates of change of CaCO_3 conversion to nitrate as monitored by ex-situ SEM-EDX analysis.
- (o) HNO_3 (initial concentration $\approx 10^{11}\text{-}10^{12}$ molecule cm^{-3}) detected using radioactive labelling (H^{13}NO_3). Arizona dust aerosol (diameter < 800 nm) was introduced into reactor via dry dispersion from its powder. Both gas and particle phase HNO_3 was detected. The efficiency of uptake of HNO_3 and its time evolution was observed to depend on the relative humidity (6-60 %) and the HNO_3 concentration. An uptake mechanism involving Langmuir like adsorption and surface reaction was proposed to explain this and derive time dependent values of γ . Data were consistent with a surface accommodation coefficient of 1.

Preferred Values

Parameter	Value	T/K
α_s	1	298
τ_{des} / s	0.1	298
$[Y] / \text{molecule cm}^{-2}$ (RH \leq 60%)	$6.5 \times 10^{13} + 4.1 \times 10^{10} * \text{RH} + 4.0 \times 10^{11} *$ RH ²	298
$k_s / \text{cm}^2 \text{ s}^{-1}$	4×10^{-15}	298
$K_{\text{LangC}} / \text{cm}^3 \text{ molec}^{-1}$	2.25×10^{-12}	298

Reliability

see comments on preferred values

Comments on Preferred Values

The experimental investigations of the kinetics of HNO₃ uptake to substrates representing atmospheric mineral aerosol show a great variability in the values of γ that have been obtained. There are several reasons for this, which are outlined below:

The role of internal surfaces in bulk, powder substrates has been assessed in different ways. Experimental uptake coefficients in Knudsen reactors are initially analysed using the geometric surface area of the sample surface (γ_{geom}). Diffusion into the sample's interstitial space on the time scale of the measurement necessitates a correction factor, which can be empirically derived from observations of a sample mass dependence of γ_{geom} . In some instances such a behaviour has been observed (Goodman et al., 2000, 2001; Underwood et al., 2001a,b; Frinak et al., 2004) and in other cases it was not observed (Fenter et al., 1995; Hanisch and Crowley, 2001a,b; Seisel et al., 2004). The uptake coefficients which use either a pore-diffusion correction or the BET surface area are orders of magnitude smaller than those relying on the geometric surface area, the correction factor depending (for pore diffusion) on the value of (γ_{geom}) obtained. In those cases where uncorrected values of γ_{geom} are available comparison shows that even here substantial deviations exist. For example, the maximum value of γ_{geom} obtained by Johnson et al. (2004) for HNO₃ uptake to CaCO₃ is still a factor of 6 lower than that obtained by Hanisch and Crowley (2001a). The influence of saturation effects (requiring correction factors of between 5 and 60) is a possible explanation as discussed by Underwood et al., (2001b). Hanisch and Crowley (2001b) have also indicated how use of high HNO₃ concentrations and reactive sample supports can result in underestimation of γ_{geom} . As shown in the Table above, results within the same research group, and using the same method and sample type reveal variations of more than two orders of magnitude in the value of γ obtained (Underwood et al, 2000; Goodman et al., 2000; Johnson et al., 2005). Thus, whereas γ_{geom} must be considered an upper limit to the true value of γ , values based on pore diffusion corrections or BET surface areas are often lower limits.

Part of the variability in γ_{geom} for any given substrate is also related to the use of differently prepared (different water content due to non-identical heating and vacuum treatments) and intrinsically different samples (e.g. Saharan sand has a very different mineralogy and alkalinity to Saharan loess). The Knudsen reactor experiments with which the majority of the kinetic data have been obtained are necessarily conducted at low humidity. For some substrates (e.g. CaCO₃) there is strong evidence that the availability of H₂O can influence the uptake coefficient (Goodman et al., 2000, 2001; Hanisch and Crowley 2001a, Vlasenko et al., 2005) whereas for others the effects of substrate heating are negligible, suggesting that sufficient strongly bound surface water is available to support high values of the initial uptake coefficient, even though the capacity may be reduced e.g. (Hanisch and Crowley, 2001a,b for uptake to Saharan and Chinese dust, Seisel et al., (2004) for uptake to α -Al₂O₃).

The aerosol flow tube experiments of Vlasenko et al. (2006, 2009) provide uptake coefficients that avoid the issues related to effective surface area calculations for bulk surfaces discussed above. They also allow variation of the relative humidity to regions found in the atmosphere. Their data indicate initial uptake coefficients of the order of 0.1, but also show that this value may change as reactive sites are consumed. Vlasenko et al. (2009) propose a parameterisation for the uptake of HNO₃ to the surface of Arizona dust, which breaks γ into the individual steps (accommodation, reversible desorption, surface reaction) and takes into account the role of RH. We have adopted their values (tabulated above) for use in the following expressions:

$$\frac{1}{\gamma} = \frac{1}{\alpha_s} + \frac{1}{\gamma_{LH}} \text{ where}$$

$$\gamma_{LH} = \frac{4k_s[Y]_s K_{LangC}(X)N_{max}}{\bar{c}(1 + K_{LangC}(X)[X]_g)} = \frac{k_s[Y]_s \alpha_s \tau_{des}}{(1 + K_{LangC}(X)[X]_g)}$$

and \bar{c} (mean thermal velocity of HNO₃) = 30000 cm s⁻¹. The RH dependence of $[Y]_s$ was derived by fitting a polynomial to their data at 6, 33 and 60 % RH and should not be extrapolated above this range. This parameterisation results in values of γ that vary from 0.03 at 6 % RH to 0.6 at 60 % RH.

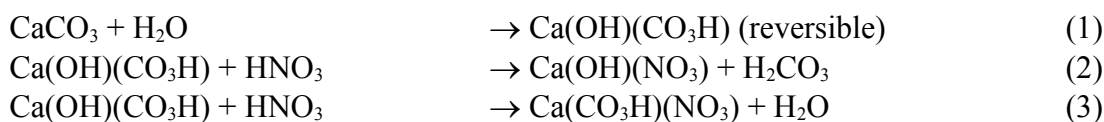
The Vlasenko et al. data appear to be the most reliable, however, when applying their numbers to atmospheric dust, some considerations must be made:

- 1) The Arizona test dust examined by Vlasenko et al. (2005) has been found to be less reactive than Saharan or Chinese dust (see Table above), which provide the bulk of emissions to the atmosphere.
- 2) We note that relative humidity will play a role, but this may be different for Saharan or Chinese dust, when compared with the CaCO₃ or Arizona dust samples investigated by Vlasenko et al. (2005). Mashburn et al. (2006) found a very strong dependence of the HNO₃ uptake to Na-montmorillonite (a clay mineral with a large capacity to adsorb water) on relative humidity. Liu et al (2008) also found a pronounced increase in HNO₃ uptake to CaCO₃ when the RH was increased from 10 to 80 %.
- 3) A time independent value of γ will not always be appropriate to model dust plumes that will age chemically during transport through the atmosphere (Vlasenko et al., 2009). This is especially the case if the uptake is limited to the surface of the particle.
- 4) For interactions where the uptake is not limited to the surface (i.e. when dealing with alkaline particles) the capacity of mineral dust to consume HNO₃ in the atmosphere will be constrained by the fraction of calcite and dolomite present. The kinetics of the uptake at long time scales may then be determined not by a surface uptake coefficient, but by the rate of dissolution of an alkaline core covered by a deliquesced, aqueous nitrate layer. There is strong laboratory evidence (Goodman et al., 2000; Mashburn et al., 2006; Prince et al., 2007) that the reaction of HNO₃ on mineral samples is not limited to the surface if sufficient H₂O is present. At exposure to high concentrations of HNO₃ at sufficient relative humidity morphological and phase changes in individual particles have been observed, with up to 40 % of particle mass converted to nitrate (Krueger et al., 2003, 2004; Laskin et al., 2005). At sufficient RH (greater than \approx 10 - 20 %) the large hygroscopicity of surface nitrate induces further HNO₃ and water uptake (Liu et al., 2008) until the particle alkalinity is neutralised. Liu et al observed a jump in the reactive uptake between 10 and 20 % RH, which they ascribe to Ca(NO₃)₂ deliquescence. HNO₃ uptake to CaCO₃ particles at high RH therefore represents a situation where the uptake can be enhanced by a chemical and thermodynamic change in the surface state. At low [HNO₃] and humidity, this is not observed (Hanisch and Crowley, 2001a) and the reaction is largely limited to the surface yet the H₂O-induced reactivation of chemically aged “dry” dust samples has been documented on several occasions (Hanisch and Crowley, 2001a; Seisel et al, 2004).

5) There are indications that dust samples from different source regions may also display different reactivity to HNO₃ (Krueger et al., 2004) so that a single value is not appropriate.

There is general agreement that the reaction of HNO₃ with mineral dust is essentially irreversible and that the products are surface and bulk nitrate species as observed spectroscopically (Goodmann et al., 2000, 2001; Börensen et al., 2000; Krueger et al., 2003, 2004; Seisel et al., 2004; Laskin et al., 2005; Mashburn et al., 2006) and also gas phase CO₂ and H₂O (Fenter et al., 1995, Goodmann et al., 2000; Hanisch and Crowley 2001a; Prince et al., 2007). Angelini et al. (2007) observed both molecularly adsorbed HNO₃ and nitrate on kaolinite and pyrophyllite samples. The reaction of HNO₃ with kaolinite was observed to generate surface adsorbed H₂O.

Mechanisms for the reaction of HNO₃ with CaCO₃ at low RH have been proposed (Al-Hosney and Grassian, 2005; Johnson et al., 2005; Santschi and Rossi, 2006) that invoke the intermediacy of Ca(OH)(CO₃H). The models attempt to explain the observed dependence of the CO₂ yield on available H₂O (Fenter, 1995, Hanisch and Crowley, 2001a, Santschi and Rossi 2006), the differences in powdered CaCO₃ and marble samples (Santschi and Rossi, 2006) and the concurrent removal of surface OH groups and formation of nitrate groups during exposure (Börensen et al., 2000; Seisel et al., 2004).



The observation of Santschi and Rossi (2006) of no CO₂ release for marble samples, yet observation of 100 % yield of H₂O led them to propose that reaction (3) dominates and that CO₂ is generated in a later step in the reaction of Ca(CO₃H)(NO₃) with a further molecule of HNO₃.

At large RH, the overall mechanism for calcite rich particles may resemble the acid-assisted, aqueous phase dissolution of carbonate, with the reaction proceeding until alkalinity is neutralised.



The uptake of HNO₃ to dust surfaces is not sensitive to chemical aging of the dust by atmospheric O₃ concentrations, at least at low RH (Hanisch and Crowley, 2003).

The time dependent γ formalism of Vlasenko et al. (2009) is appropriate for those time scales in which the processing of alkaline mineral dust particles by HNO₃ is restricted to the surface only. Both field observations of large nitrate content of mineral dust particles (e.g. Matsuki et al., 2005) and the laboratory observations made at high RH suggest that for the time scales associated with dust transport lifetimes (days), the uptake of HNO₃ will be controlled by rates of dissolution of alkaline components in an aqueous layer.

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