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Study of the pressure dependence of the rate constant of the reaction of NO_3 with NO_2

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ABSTRACT

We have applied time resolved cavity ring-down spectroscopy to a kinetic study of the reaction of NO_3 with NO_2 in 20–700 Torr of N_2 diluent at 298 and 283 K. By fitting the data to Troe's expression with $F_c = 0.6$, the low-pressure and high-pressure limiting rate constants of this reaction were determined to be $k_1^{low}(298\ K) = (3.1 \pm 0.8) \times 10^{-30}$ cm^6 $molecule^{-2}$ s^{-1} and $k_1^{high}(298\ K) = (1.6 \pm 0.3) \times 10^{-12}$ cm^3 $molecule^{-1}$ s^{-1}. These rate constants thus determined were 10–40% larger than those recommend in the NASA/JPL database.

1. Introduction

Nitrogen trioxide, i.e., nitrate radical NO_3, is considered to be one of the most important oxidizers in urban areas, because NO_3 is formed by the reaction of ozone (O_3) with nitrogen dioxide (NO_2) included in automobile exhaust emissions. Solar photolysis suppresses the concentration of NO_3 in the daytime resulting in the mixing ratio of NO_3 peaking at nighttime. The NO_3 radical can reversibly combine with NO_2 to form dinitrogen pentoxide (N_2O_5) by the following equilibrium reaction (1,-1):

$$NO_3 + NO_2 + M \rightleftarrows N_2O_5 + M, \qquad (1,\text{-}1)$$

where M implies third bodies [1–10]. This reaction is one of the chemical reactions that play important roles on chemical transformation in the earth's atmosphere and the oxidative capacity of the troposphere. An atmospheric N_2O_5 formed in reaction (1) can react heterogeneously with water to give two molecules of nitric acid (HNO_3) which is a longer-lived reservoir species, and N_2O_5 can be removed from the atmosphere via wet or dry deposition [11]. Thus, the equilibrium constant of reaction (1,-1), i.e. the ratio of the rate constants of reactions (1) to (-1), is important in determining the oxidative capacity of the troposphere.

Because reaction (1) is a termolecular reaction, its rate constant is usually characterized in accord with Troe's expression (2):

$$k_1 = \left(\frac{k_1^{low}[M]}{1 + \left(k_1^{low}[M]/k_1^{high} \right)} \right) F_c \left\{ 1 + \left[\log \left(k_1^{low}[M]/k_1^{high} \right) \right]^2 \right\}^{-1}, \qquad (2)$$

where k_1^{low} and k_1^{high} are low-pressure and high-pressure limiting rate constants of reaction (1), respectively. The constant of F_c is the broadening factor, the value of which may be different by each reaction. In the NASA/JPL database, the value was fixed to $F_c = 0.6$ in analyzing the rate constants for reaction (1) reported previously. On the other hand, the value of $F_c = 0.35$ was used in the IUPAC data base in analyzing the different set of rate constants for reaction (1). Consequently, we can see the difference between the recommendation values of k_1^{low}, k_1^{high} and F_c by NASA/JPL [1] and IUPAC [2].

In 2000, Burkholder and Ravishankara reported the rate constant for the reaction of $O(^3P) + NO_2$ in 20–800 Torr of N_2 diluent [12]. To determine the rate constant, they monitored the time profiles of NO_3 concentration after the 355-nm pulse laser photolysis of NO_2 in NO_2/N_2 gas mixture by using the transient long-path 662-nm diode laser absorption method. In their reaction system, reaction (1) affected the loss of NO_3 formed in the reaction of $O(^3P) + NO_2$. In analyzing the data, they included an additional NO_3 first-order loss rate coefficient which depended on $[NO_2]$ to reproduce the fast loss of NO_3. They suggested that the increase of the k_1 value by 10–40% from those recommend by NASA/JPL would have nearly the same effect as introducing an additional rate coefficient for NO_3 first-order loss. This suggestion may indicate that the value of k_1 recommend by NASA/JPL is underestimated by 10–40% in the total pressure of 20–800 Torr.

We confirm the results of Burkholder and Ravishankara through the experiment of 355-nm pulse laser photolysis of NO_2 in NO_2/N_2 gas mixture using time-resolved cavity ring-down spectroscopy (TR-CRDS). In this study, we determine the rate constant of reaction (1) in 20–700 Torr of total pressure using TR-CRDS, and reveal the underestimation of k_1 in the NASA/JPL data base. Because of the importance of reaction (1) in the atmospheric chemistry, the results obtained in this work are valuable for model simulations.

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2. Experimental

The TR-CRDS apparatus used in the present study is similar to that reported previously [13,14]. Two pulsed lasers were employed. A Nd^{3+}: YAG laser (Continuum Co., Surelite II) was used to generate the NO_3 radicals in the photolysis of N_2O_5 at 266 nm. A dye laser (Sirah Co., Cobra-Stretch; DCM Dye) pumped by the 532 nm output of a Nd^{3+}: YAG laser (Continuum Co., Surelite I) was used to probe the concentration of NO_3 at 662 nm. After the photolysis laser beam traversed a reaction cell nearly collinear to the axis of the ring-down cavity, the probe laser beam was injected through one of two high-reflectivity mirrors that made up the ring-down cavity. The mirrors (Research Electro Optics) mounted 1.04 m apart had a specified maximum reflectivity of 0.999 at 635 nm, a diameter of 7.75 mm, and a radius of curvature of 1 m. Light leaking from one of the mirrors of the ring-down cavity was detected by a photomultiplier (Hamamatsu Photonics Co., R928) through a broad band pass filter (656 nm, FWHM 10 nm). The decay of the light intensity was recorded using a digital oscilloscope (Tektronix, TDS430A) and transferred to a personal computer. The decay of the light intensity is given by equation:

$$I(t) = I_0\exp(-t/\tau) = I_0\exp\{-t/\tau_0 - \sigma nc(L_R/L)t\}, \tag{3}$$

where $I(t)$ is the intensity of light at time t; τ_0 is the cavity ring-down time (~10 μs at 662.0 nm) without photolysis laser light; L_R is the length of the reaction region (0.46 m) while L is the cavity length (1.04 m); τ is the cavity ring-down time with photolysis light; σ and n are the absorption cross section and concentration of the species of interest, respectively; c is the speed of light.

The reaction cell consisting of a Pyrex glass tube (21 mm i.d.) was evacuated by an oil rotary pump through a liquid N_2 trap. The volume of the detection region in the reaction cell was 160 cm^3. The temperature of the gas flow region was controlled by the circulation of thermostated water and kept at 298 or 283 K. The difference between the temperatures of the sample gas at the entrance and exit of the flow region was <0.1 K. The pressure in the cell was monitored by an absolute pressure gauge (MKS, Baratron). Gas flow rates were regulated and measured by mass flow controllers (KOFLOC, Model 3660 and Model 3440). A slow flow of nitrogen diluent gas was introduced at both ends of the ring-down cavity close to the mirrors to minimize the deterioration caused by exposure to reactants and products. The total flow rate was kept constant at 127 $cm^3\ s^{-1}$. Experiments were performed with 1 Hz laser operation under the conditions that the gas mixture in the detection region was replenished every 1–2 laser shots.

The mixtures of N_2O_5/N_2 and NO_2/N_2 were prepared and stored in glass bulbs, and were injected into the reaction cell using mass flow controllers. The concentrations of N_2O_5 and NO_2 in the reaction cell could be calculated by using the flow rates.

We prepared N_2O_5 in the dehydration of HNO_3 in the presence of P_2O_5 as described previously [13,15]. Gaseous NO_2 was produced by the reaction of NO with excess O_2. The $NO_2/NO/O_2$ gas mixture was passed through a trap at methanol slash temperature, and the collected NO_2 was purified by a trap-to-trap distillation until a pure white solid was obtained. Other reagents were obtained from commercial sources. NO (99%), N_2 (>99.9995%) and O_2 (>99.9995%) were used without further purification.

3. Results and discussion

We performed some experiments where the NO_3 absorption-time profiles were monitored after the 355-nm photolysis of NO_2 in NO_2/N_2 gas mixture using TR-CRDS. In these experiments, we observed the same phenomena as reported by Burkholder and Ravishankara [12]; the 10–40% larger values of k_1 as compared with those recommended by NASA/JPL were necessary to reproduce the fast loss of NO_3. There are some difficulties for the determination of the rate constant of reaction (1) in NO_2/N_2/355 nm experiments because of the existence of the many influences by sub-reactions.

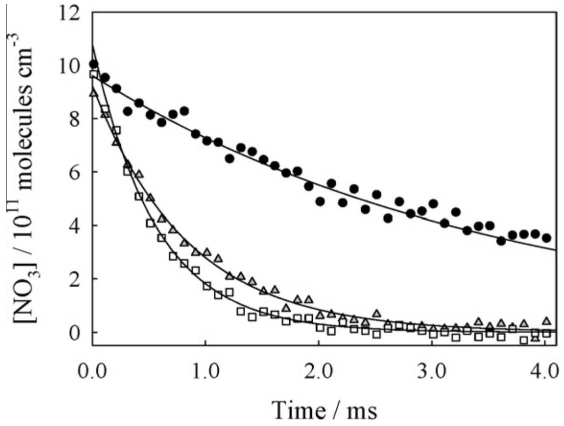

Figure 1. Typical decay profiles of NO_3 in 100 Torr of N_2 diluent at 298 K. $[NO_2]_{add} = 0$ (circles), 9.7×10^{14} (triangles) and 1.6×10^{15} molecules cm^{-3} (squares). The solid curves are the fits of the data to Eq. (5).

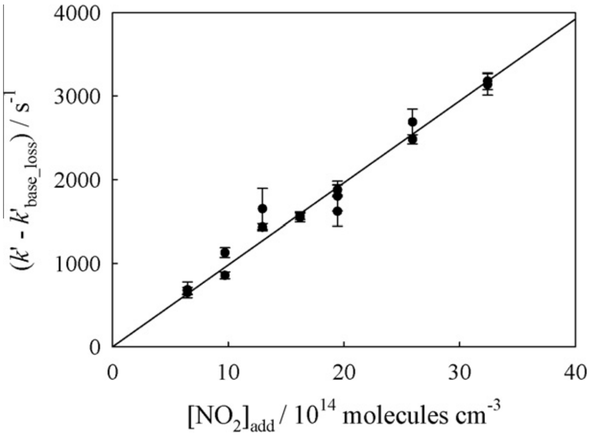

Figure 2. Plot of $(k' - k'_{base\_loss})$ vs. $[NO_2]_{add}$ in 100 Torr of N_2 diluent at 298 K. The solid line is a result of the linear least squares fit.

To minimize the influences of sub-reactions, we performed the experiments, in which NO_3 absorption-time profiles were monitored after the 266-nm photolysis of N_2O_5 in $N_2O_5/NO_2/N_2$ gas mixture using TR-CRDS for determination of the rate constant reaction (1). In the N_2O_5/N_2 mixture stored in a glass bulb, N_2O_5 was gradually decomposed to NO_2 before the N_2O_5/N_2 mixture was flowed into the reaction cell. The decomposition of a N_2O_5 molecule gives two NO_2 molecules; NO_3 formed in reaction (-1) is converted to NO_2 by a self reaction, a wall reaction, and so on. To check the actual concentrations of NO_2 in a N_2O_5/N_2 mixture, we measured the IR spectra of the gas mixtures sampled from the N_2O_5/N_2 mixture in the glass bulb by the FTIR spectrometer. As the result, we experimentally determined the concentration of NO_2 and estimated the decomposition of N_2O_5 to be about 4% after 30 min and about 16% after 210 min. Following these results, we have carried out the experiments under the conditions that the concentrations of NO_2 originating from the decomposition of N_2O_5 were greater than 1×10^{14} molecules cm^{-3} in TR-CRDS reaction cell even without the deliberately-added NO_2. Here, $>1 \times 10^{14}$ molecules cm^{-3} of NO_2 are required to achieve the pseudo first order conditions where

Table 1
Pressure effect on the rate constants of $NO_3 + NO_2$ at 298 K.

Pressure (Torr)	$[N_2O_5]_0$ (10^{14} molecules cm^{-3})	$[NO_2]_{add}$ (10^{14} molecules cm^{-3})	$[NO_3]_0$ (10^{12} molecules cm^{-3})	k_1 (10^{-12} cm^3 $molecule^{-1}$ s^{-1})
20	3.3–4.9	6.5–32	1.0–1.6	0.58 ± 0.07
50	3.2–4.9	6.5–32	0.6–1.9	0.81 ± 0.09
100	2.1–8.3	6.5–32	0.4–1.5	0.98 ± 0.11
200	3.2–6.6	3.2–65	1.0–2.1	1.20 ± 0.13
300	1.6–3.2	3.2–16	0.4–1.2	1.23 ± 0.15
400	1.6–3.2	3.2–16	0.5–1.2	1.32 ± 0.15
500	3.2	3.2–16	0.8–1.4	1.33 ± 0.14
700	6.8	4.5–14	2.3–2.6	1.43 ± 0.17

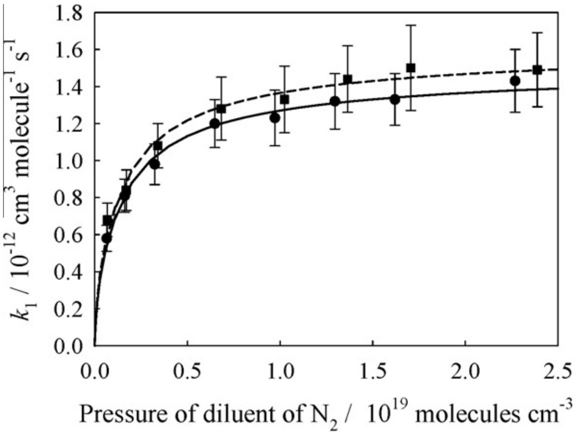

Figure 3. Plot of rate constants of NO_3 with NO_2 as a function of N_2 diluent pressure at 298 K (circles) and 283 K (squares). The solid curve and dashed curve are the fits of the data at 298 and 283 K to Eq. (2), respectively.

the NO_3 decay profiles can be fitted by a single exponential decay without the deliberately-added NO_2.

Then, we started to measure the temporal concentration of NO_3 radicals with TR-CRDS by monitoring the electronic transition NO_3 ($B^2E' \leftarrow X^2A_2'$) at 662.0 nm. The absorption of NO_3 was converted to the concentration by using the reported absorption cross section at 662 nm, $\sigma_{NO3}(662\ nm) = 2.25 \times 10^{-17}\ cm^2\ molecule^{-1}$ [1,16,17]. The NO_3 radicals were produced by the 266 nm photolysis of N_2O_5 with the concentration of $(2.1–8.3) \times 10^{14}$ molecules cm^{-3} in 100 Torr of N_2.

$$N_2O_5 + h\nu(266\,nm) \rightarrow NO_3 + NO_2 \qquad (4a)$$

$$\rightarrow NO_3 + NO + O(^3P) \qquad (4b)$$

The value of $\sigma_{N2O5}(266\ nm)$ was reported to be $2.0 \times 10^{-19}\ cm^2\ molecule^{-1}$ [1]. The quantum yields of reactions (4a) and (4b) were reported to be 0.62 and 0.38, respectively [2,18]. A typical decay profile of NO_3 generated by the 266-nm photolysis of mixtures of N_2O_5/N_2 in the reaction cell at 298 K is shown by closed circle in Figure 1. The NO_3 decay profile shows a single exponential decay where the NO_3 is consumed with about 3.8 ms of lifetime. This NO_3 loss is considered to result from the reaction with NO_2 which are produced by the decomposition of N_2O_5 in the glass bulb. The concentration of NO_2 form N_2O_5 in the reaction cell was $>1.0 \times 10^{14}$ molecules cm^{-3} from the results of FTIR experiments as shown above, which was high enough to give the exponential decay of NO_3. Because the concentrations of the sub-products in reaction (4), such as NO_2, NO and $O(^3P)$, are three orders of magnitude less than that of NO_2 injected as the contamination from the N_2O_5/N_2 mixture in the glass bulb, the photolytically-generated NO_2, NO and $O(^3P)$ do not have an influence on the loss of NO_3 radical. The influence of reaction (-1) on the NO_3 loss could be neglected for observed NO_3 decay because of the small value of the rate constant of reaction (-1) ($k_{-1} = 0.046\ s^{-1}$ at 298 K) [19].

To determine the rate constant of the reaction of NO_3 with NO_2, temporal profiles of NO_3 were measured with deliberately-added NO_2 ($[NO_2]_{add} = 3.2–32 \times 10^{14}$ molecules cm^{-3}) by the introduction of a gas mixture of NO_2/N_2 prepared in a glass bulb. Here, $[NO_2]_{add}$ is so much in excess of the NO_3 concentration. The NO_3 decay could be fitted by Eq. (5):

$$[NO_3]_t = [NO_3]_0 \exp(-k't). \qquad (5)$$

In Eq. (5), k' is expressed by the following equation:

$$k' = k_1[NO_2]_{add} + k'_{base\_loss}, \qquad (6)$$

where k'_{base_loss} is the sum of first order decay components such as the reaction with NO_2 produced by the decomposition of N_2O_5 in N_2O_5/N_2 glass bulb, wall loss and diffusion form the detection region. As described above, k'_{base_loss} under our experimental conditions consists mainly of the reaction of NO_3 with NO_2 which are produced by the decomposition of N_2O_5 in the glass bulb. Here, we can expect that $O(^3P)$ and NO produced from the 266-nm photolysis of NO_2 do not affect the observed NO_3 decays. By using absorption cross sections of NO_2 and N_2O_5, at 266 nm, the concentrations of $O(^3P)$ and NO were estimated to be $<1.5 \times 10^{11}$ molecules cm^{-3} under our experimental conditions. These concentrations are about 3–4 orders of magnitude less than that of NO_2. The rate constants of the reaction of NO_3 with $O(^3P)$ and NO is reported to be $1.0 \times 10^{-11}\ cm^3\ molecule^{-1}\ s^{-1}$ and $2.6 \times 10^{-11}\ cm^3\ molecule^{-1}\ s^{-1}$, respectively [1]. By considering the concentrations of $O(^3P)$ and NO and their reactivity toward NO_3, it is safe to say that the influence of O and NO on the determination of the rate constant of $NO_3 + NO_2$ could be ignored. The value of k'_{base_loss} could be determined by the least-squares analysis of NO_3 decay profile measured with $[NO_2]_{add} = 0$ with Eqs. (5)

Table 2
Summary of the reported low-pressure and high-pressure limiting rate constants of the rate constants of $NO_3 + NO_2$ at 298 K.

k_1^{low} (10^{-30} cm^6 $molecule^{-2}$ s^{-1})	k_1^{high} (10^{-12} cm^3 $molecule^{-1}$ s^{-1})	F_c	Temperature (K)	Reference
2.0	1.4	0.60	298	NASA/JPL recommendation, 2006 [1]
3.6	1.9	0.35	298	IUPAC recommendation, 2006 [2]
3.1	1.6	0.60	298	This work
3.3	1.7	0.60	283	This work

Table 3
Pressure effect on the rate constants of $NO_3 + NO_2$ at 283 K.

Pressure (Torr)	$[N_2O_5]_0$ (10^{14} molecules cm^{-3})	$[NO_2]_{add}$ (10^{14} molecules cm^{-3})	$[NO_3]_0$ (10^{12} molecules cm^{-3})	k_1 (10^{-12} cm^3 molecule^{-1} s^{-1})
20	3.4–6.8	4.7–23	0.4–1.0	0.68 ± 0.09
50	6.6–6.8	4.9–25	0.5–0.8	0.84 ± 0.11
100	6.8	5.1–26	0.6–0.8	1.08 ± 0.12
200	10	10–30	0.9–1.8	1.28 ± 0.17
300	6.8	3.4–10	1.5–1.7	1.33 ± 0.18
400	6.8	3.4–10	1.8–1.9	1.44 ± 0.18
500	6.8	3.4–10	1.1–1.4	1.50 ± 0.23
700	9.6	4.8–15	2.8–3.0	1.49 ± 0.20

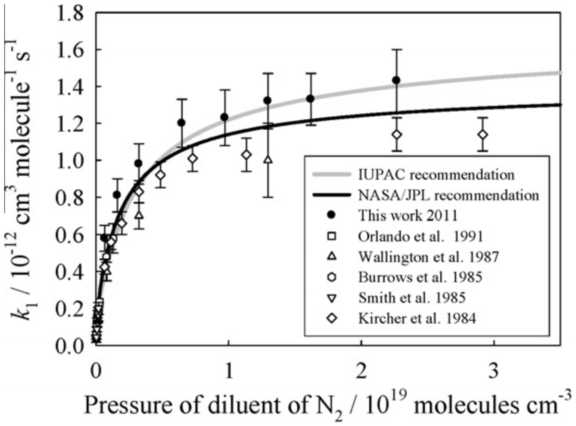

Figure 4. Plot of reported rate constants of NO_3 with NO_2 as a function of N_2 diluent pressure at 298 K. The data of this work (closed circles), Kircher et al. (open diamonds), Smith et al. (opened down triangles), Burrows et al. (open hexagons), Wallington et al. (opened up triangles) and Orlando et al. (opened squares) are shown. The black curve and gray curve are the falloff curves of NASA/JPL and IUPAC recommendations at 298 K, respectively.

and (6). As discussed above, the value of k'_{base_loss} gradually increased during experiments because the N_2O_5 molecules in glass bulb gradually decomposed to NO_2 during the experiments. We measured k' with $[NO_2]_{add} = 0$ before and after the measurements with $[NO_2]_{add} \neq 0$, and corrected the change of k'_{base_loss} in determining the rate constant of reaction (1). By this procedure, the term of k_1 $[NO_2]_{add}$ in Eq. (6) could be extracted with reasonable accuracy. Figure 2 shows ($k' - k'_{base\_loss}$) vs. $[NO_2]_{add}$ in 100 Torr of N_2 diluent at 298 K. By considering the relationship of Eq. (5), a linear least-squares analysis gives $k_1 = (0.98 \pm 0.11) \times 10^{-12}$ cm^3 molecule^{-1} s^{-1}. The quoted uncertainty is derived by considering the uncertainties of calibrations of $[NO_2]_{add}$, measurements of pressure, mass flow rates, and reaction path length, and also the fitting errors.

To test the pressure dependence of a diluent gas on k_1, additional experiments were performed in 20, 50, 100, 200, 300, 400, 500 and 700 Torr of N_2 diluent at 298 K. As summarized in Table 1, we determined k_1 to be $(0.58 \pm 0.07) \times 10^{-12}$ in 20 Torr, $(0.81 \pm 0.09) \times 10^{-12}$ in 50 Torr, $(0.98 \pm 0.11) \times 10^{-12}$ in 100 Torr, $(1.20 \pm 0.13) \times 10^{-12}$ in 200 Torr, $(1.23 \pm 0.15) \times 10^{-12}$ in 300 Torr, $(1.32 \pm 0.15) \times 10^{-12}$, in 400 Torr $(1.33 \pm 0.14) \times 10^{-12}$ in 500 Torr and $(1.43 \pm 0.17) \times 10^{-12}$ cm^3 molecule^{-1} s^{-1} in 700 Torr of N_2 diluent. As shown in Figure 3 the data were fit to Troe's expression (2). Here the value of F_c was adopted to be 0.6 to compare with the NASA/JPL recommendation value. The values of the low-pressure and high-pressure limiting rate constants of reaction (1) were determined to be $k_1^{low}(298\text{ K}) = (3.1 \pm 0.8) \times 10^{-30}$ cm^6 molecule^{-2} s^{-1} and $k_1^{high}(298\text{ K}) = (1.6 \pm 0.3) \times 10^{-12}$ cm^3 molecule^{-1} s^{-1}. These results are summarized in Table 2 with those values of NASA/JPL and IUPAC recommendations. Additionally, k_1 in 20–700 Torr of N_2 diluent at 283 K were measured and are summarized in Table 3. By the fitting of the data to Troe's expression (2) with $F_c = 0.6$, The values for the low-pressure and high-pressure limiting rate constants of reaction (1) at 283 K were also determined to be $k_1^{low}(283\text{ K}) = (3.4 \pm 1.0) \times 10^{-30}$ cm^6 molecule^{-2} s^{-1} and $k_1^{high}(298\text{ K}) = (1.7 \pm 0.3) \times 10^{-12}$ cm^3 molecule^{-1} s^{-1}. These results are also summarized in Table 2. For comparison, the data of k_1 at 298 K reported by Kircher et al. [5], Smith et al. [6], Burrows et al. [7], Wallington et al. [8] and Orlando et al. [9], which were used to obtained k_1^{low} and k_1^{high} of NASA/JPL recommendation, are shown with ours in Figure 4. The falloff curves of NASA/JPL and IUPAC recommendations are also indicated in Figure 4. As shown in Figure 4, the values of k_1 determined in this work are in excellent agreement with the falloff curve of IUPAC recommendations and some of present values are close to some of those reported previously. The values of k_1 determined in this work are 10–40% larger than those recommend by NASA/JPL [1]. This result is consistent with the suggestion by Burkholder and Ravishankara in their paper [12]. Because of the importance of reaction of NO_3 with NO_2 in the atmospheric chemistry, the results of this work are considered to be valuable for atmospheric model simulations.

Acknowledgments

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