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AE REACTION RATE DATA

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Final Report

July 1 - December 31, 1978

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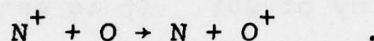
Abstract A summary is presented of the ionospheric rate coefficients that are derived from Atmosphere Explorer data taking into account laboratory and theoretical studies. A rate coefficient of  $2.2 \times 10^{-12} \text{ cm}^3 \text{ s}^{-1}$  has been derived for the reaction  $\text{N}^+ + \text{O} \rightarrow \text{N} + \text{O}^+$  at about 840K, with a probable accuracy of 20%. It is established that the reaction  $\text{O}^+ + \text{N}(^2\text{D}) \rightarrow \text{O} + \text{N}^+$  is a significant source of  $\text{N}^+$  ions. The corresponding rate coefficient is about  $1.3 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ . The associative ionization reaction  $\text{N}(^2\text{D}) + \text{O} \rightarrow \text{NO}^+ + \text{e}$  is not important in the undisturbed atmosphere.

1. Introduction

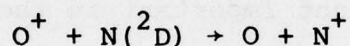
The Atmosphere Explorer satellites carried a complement of instruments to measure simultaneously the ion and neutral composition of the atmosphere, the electron and neutral particle temperatures, various airglow emissions and the solar ionizing flux (Dalgarno et al. 1973). Detailed theoretical analyses have been performed from which empirical values of the rate coefficients of atmospheric ion-molecule reactions have been derived. The AE results have been reviewed recently (Torr and Torr 1978, Swider 1978). Laboratory studies have continued and they have also been reviewed recently (Albritton 1978, Ferguson et al. 1979).

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We present here a critical comparison of the atmospheric and laboratory data and we report on the results of our analysis of AE data that has led to an empirical value of the rate coefficient for the charge transfer reaction



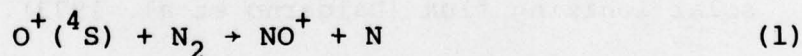
We introduce as a new source of  $N^+$  the reaction



and show that it is capable of explaining the AE composition data on  $N^+$ .

## 2. Reaction rate coefficients

### 2.1 Ground state $O^+(^4S)$ ions



A detailed discussion of the rate coefficient,  $k_1$ , of reaction (1) has been given by Torr and Torr (1978). The rate coefficient depends upon the vibrational population of  $N_2$  and on the ion temperature  $T_i$ . At 300K, laboratory measurements (cf. Albritton et al. 1977, Smith et al. 1978,

Glosik et al. 1978, Chen et al. 1978) all now agree that  $k_1 = 1.2 \times 10^{-12} \text{ cm}^3 \text{ s}^{-1}$ . Using a heated drift-tube mass spectrometer apparatus in which the vibrational population of  $\text{N}_2$  is characterized by the kinetic temperature of the gas and ions, Chen et al. (1978) have measured  $k_1$  up to 900K obtaining there a value of  $0.5 \times 10^{-12} \text{ cm}^3 \text{ s}^{-1}$ . The values of  $k_1$  tend to be smaller than the heated afterglow results of Lindinger et al. (1974), though within the large scatter of the afterglow data, but they agree well with the flow-drift data of Albritton et al. (1977). In the flow-drift measurements, the vibrational population corresponds to 300 K and only the ion temperature is elevated. The agreement between the measurements of Chen et al. and Albritton et al. suggests that vibrational populations corresponding to temperatures less than 900 K have little effect on the rate coefficients. The AE analyses give reliable information only at an ion temperature of about 750 K and the derived rate coefficient is in harmony with the measured value. St.-Maurice and Torr (1978) have used the data of Albritton et al. (1977) to calculate  $k_1$  as a function of T.

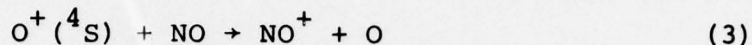
The measurements of Albritton et al. (1977) show that a rapid increase in the rate coefficient occurs at temperatures above about 1000 K. It is not yet clear how much of the increase is due to the increasing population of the higher vibrational levels but it seems likely that the rate coefficient

for reaction of  $O^+$  with  $N_2$  in a particular vibrational level does increase with increasing velocity after passing through a minimum. In atmospheric conditions, the ion velocity distribution may not be Maxwellian (cf. St.-Maurice and Schunk 1979). St.-Maurice and Torr (1978) have drawn attention to the sensitivity of  $k_1$  to departures from Maxwellian.

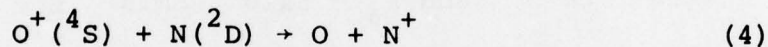


At 300 K, laboratory data are now all consistent with a rate coefficient  $k_2$  of  $2.1 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$ . In their experiments in which thermal equilibrium characterizes the vibrational populations, Chen et al. (1978) measured a slow decrease of  $k_2$  with increasing T to a value of  $1.0 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  at T=700 K. Their results agree closely with the heated afterglow values of Lindinger et al. (1974) but are systematically less by the order of 30% than the flow-drift data of Albritton et al. (1977). According to Chen et al. (1978), the discrepancy suggests either that the rate coefficient decreases with increasing vibrational excitation or that the conversion of the flow-drift cross section data to thermal rate coefficients is inaccurate. St.-Maurice and Torr (1978) have studied the effect on  $k_2$  of different ion-velocity distributions.

Analyses of AE data are not sufficiently accurate to distinguish between the two sets of measurements and are consistent within the uncertainties with both.

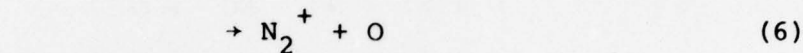
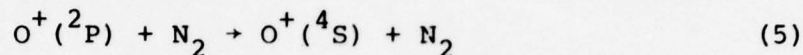


AE analyses have not given any estimates of  $k_3$ . St.-Maurice and Torr (1978) have used cross sections measured by Albritton et al. (1977) to calculate  $k_3$  as a function of T.

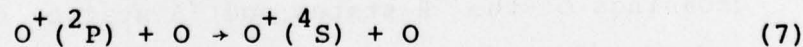


A comprehensive analysis of the AE data on  $\text{N}^+$  that we have performed has established that reaction (4) is a major source of  $\text{N}^+$  ions. We have derived a rate coefficient  $k_4$  of  $1.3 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  to a probable accuracy of 50%. An examination of the potential energy curves of  $\text{NO}^+$  (Michels 1979) shows a close approach of the  ${}^5\Sigma^+$  curves dissociating into the initial and final states of reaction (4) and avoided crossings of the  ${}^3\Pi$  states and  ${}^3\Delta$  states, suggesting that the reaction will indeed be rapid.

2.2 Metastable  $O^+(^2P, ^2D)$  ions.

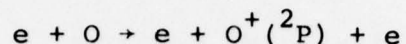


AE data on emission of the 7320-7330 Å doublet originating in the metastable  $O^+(^2P)$  level have been analyzed to yield  $k_5+k_6 = (4.8\pm 1.4)\times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  (Rusch et al. 1977). Analyses of  $N_2^+$  data (Oppenheimer et al. 1977) suggest that  $k_5 = 4\times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  and  $k_6 = 5\times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  but these separate estimates are much less accurate than is their sum. The laboratory data of Glosik et al. (1978) strongly suggest in fact that charge transfer is more likely than quenching. The large quenching coefficient  $k_5$  was required by Oppenheimer et al. to avoid too substantial a source of  $N_2^+$  at high altitudes. The discrepancy with laboratory data can be resolved by postulating a larger rate coefficient for deactivation of  $O^+(^2P)$  by atomic oxygen:

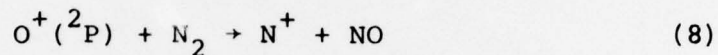


Rusch et al. (1977) derived a rate coefficient for reaction (7) of  $(5.2\pm 2.5)\times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  whereas Oppenheimer

et al. (1977) appear to require a value in excess of  $2 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ . The contribution to  $\text{O}^+(^2\text{P})$  production by photoelectron impact

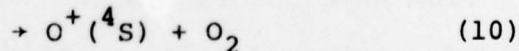
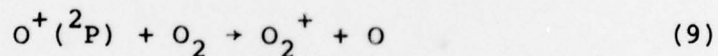


is one source of the discrepancy between the different analyses of the AE data. Alternatively  $\text{O}^+(^2\text{P})$  may undergo reaction with  $\text{N}_2$  according to the channel

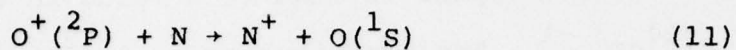


Our analysis of the  $\text{N}^+$  composition suggests a rate coefficient of  $1.3 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  though the value is not yet secure.

AE data have provided no data on the reactions of  $\text{O}^+(^2\text{P})$  with  $\text{O}_2$ :

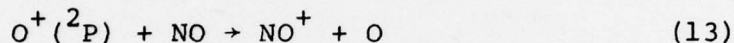


For (9), Glosik et al. (1978) measure a rate coefficient of  $1.3 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  for a mixture of  $\text{O}^+(^2\text{P})$  and  $\text{O}^+(^2\text{D})$  ions. By assuming that

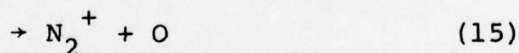
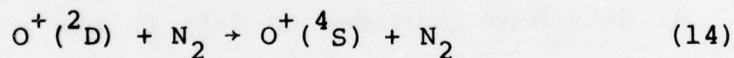


is a source of  $\text{O}(^1\text{S})$  atoms, AE observations of the given line at 5577 Å have yielded (Torr et al. 1975) a value for  $k_{11} + k_{12}$  of  $1.7 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ .

AE data provide no information on

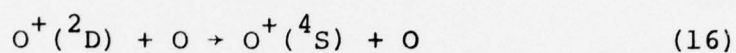


The laboratory data of Glosik et al. (1978) are consistent with  $k_{13} = 1.2 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$  though this value could be attributed to reaction with  $\text{O}^+(^2\text{D})$  ions.

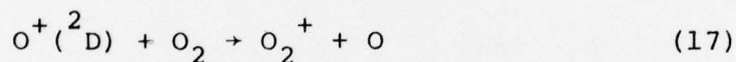


From AE-C observations of  $\text{N}_2^+$ , Torr and Orsini (1977) derived a value of  $(5 \pm 2.5) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  for  $k_{15}$ . The value is larger than the rate coefficient of  $1.4 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  measured for a mixture of  $\text{O}^+(^2\text{D})$  and  $\text{O}^+(^2\text{P})$  ions (Glosik et al. 1978). Torr and Orsini (1977) have probably underestimated the uncertainties in the theoretical analysis. Indeed a more

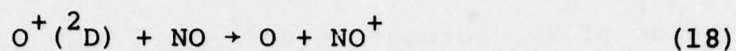
recent study of AE data by Orsini et al. (1977) suggests a value of  $k_{15}$  between  $5 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  and  $3 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ . Orsini et al. (1977) also derive a rate coefficient for reaction (14) between zero and  $0.5k_{15}$  which is within its very broad limits consistent with the value of  $1.5 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  suggested by laboratory data (Glosik et al. 1978).



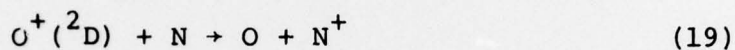
The most recent study of AE data (Torr and Orsini 1978) yields an upper limit  $k_{16} \ll 3 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  consistent with quantum mechanical considerations.



Torr et al. (1979) have argued that reaction (17) is an important source of  $\text{O}_2^+$  under certain atmospheric conditions and have derived from AE data on  $\text{O}_2^+$  composition a value of  $k_{17}$  of  $2 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ , a value which seems large in view of the laboratory data of Glosik et al. (1978) indicating  $1.3 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ . The accuracy of the empirical value of Torr et al. (1979) is probably low.

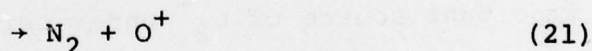
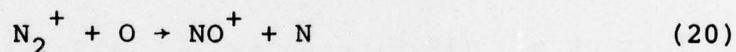


AE observations have not yielded any information on reaction (18). Glosik et al. (1978) indicate a rate coefficient of  $1.2 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$  if  $\text{O}^+(^2\text{D})$  and  $\text{O}^+(^2\text{P})$  react at a similar rate.



Our analysis of AE data on is consistent with an upper limit for  $k_{19}$  of  $3 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ . The actual value is probably much smaller.

### 2.3 $\text{N}_2^+$ ions

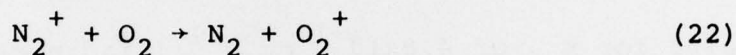


The most refined estimate of  $k_{20}$  from AE data is probably that of Torr and Orsini (1978) who find that

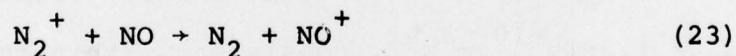
$$k_{20} = 1.4 \pm 0.3 \times 10^{-10} (\text{Ti}/300)^{-0.44} .$$

Frederick and Rusch (1977) have shown from an analysis of NI 5199 Å emission measured by AE that essentially every reaction of (20) leads to an N(<sup>2</sup>D) atom. The AE rate coefficient is consistent with laboratory data (McFarland et al. 1974). According to laboratory data,  $k_{20}/k_{21} = 0.95$ , relatively independent of T so that  $k_{21}$  may be written in the form

$$k_{21} = 7.4 \times 10^{-12} (\text{Ti}/300)^{-0.44} .$$



Reaction (22) is not very important at the altitudes of the AE satellites. The laboratory value at room temperature of  $k_{22}$  is  $5.1 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  (Smith et al. 1978).

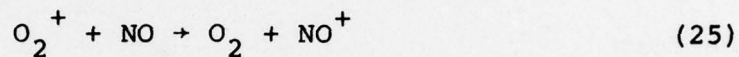


A value for  $k_{23}$  of  $3.3 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  at 300 K has been obtained from laboratory data by Fehsenfeld et al. (1970).



An upper limit to  $k_{24}$  of  $1 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  has been given by Ferguson (1968). It is consistent with our analysis of AE data on the  $\text{N}^+$  composition.

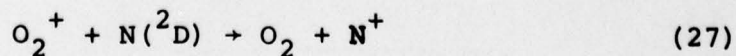
#### 2.4 Ground state $\text{O}_2^+$ ions



The rate coefficient of reaction (25) has not been derived from AE data. The AE data are consistent with the laboratory value for  $k_{25}$  of  $4.8 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  (Lindinger et al 1975, Smith et al 1978).

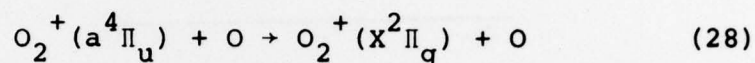


Laboratory data on reaction (26) (Fehsenfeld 1977) give  $k_{26} = 1.2 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ , consistent within the uncertainties with the rate coefficient of  $1 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  obtained from AE data by Torr et al. (1976). Frederick et al. (1976) and Kopp et al. (1977) have estimated from AE data the rate coefficient for the specific channel in (26) which produces  $\text{O}(^1\text{S})$  atoms (cf. Swider 1978).

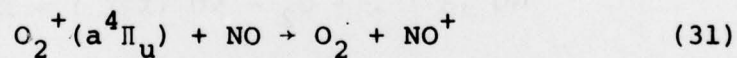
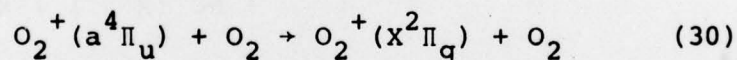
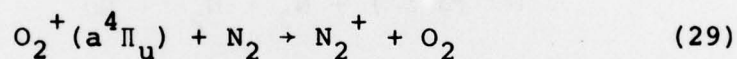


There are no laboratory data on reaction (27). It may be possible with further analysis to devise a rate coefficient from AE data. Dalgarno (1970) has suggested a value of  $4 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  but it is very uncertain.

2.5 Metastable  $\text{O}_2^+(\text{a}^4\Pi_u)$  ions.



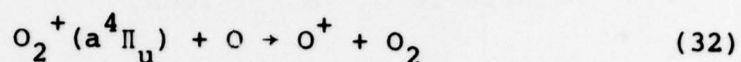
Atmosphere Explorer data are generally consistent with a rapid deactivation of  $\text{O}_2^+$  metastable ions and reveal no evidence of a participating  $\text{O}_2^+$  ion that is not in its ground state (Oppenheimer et al. 1977b). The reaction (28) may have a rate coefficient of about  $10^{-9} \text{ cm}^3 \text{ s}^{-1}$ . Laboratory measurements have been made of the reactions



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The measured rate coefficients are  $k_{29} = 4.1 \pm 1.6 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  (Lindinger et al 1975) or  $6.0 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  (Glosik et al 1978),  $k_{30} = 3.1 \pm 1.5 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  (Lindinger et al, 1975) or

$4.6 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  (Glosik et al. 1978) and  $k_{31} = 1.0 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$  (Glosik et al. 1978). Ferguson (1979) has noted the possible importance in the F region of

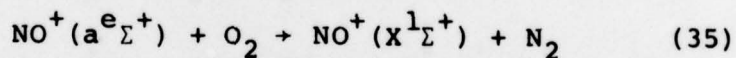
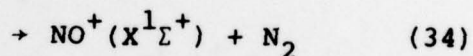
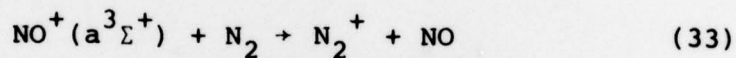


It may be possible to obtain an empirical value from AE data

### 2.6 Metastable $\text{NO}^+$ ions

Metastable  $\text{NO}^+$  ions may play a role especially in the disturbed ionosphere. AE data provide no information.

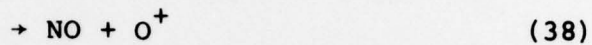
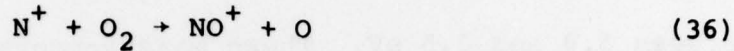
Laboratory measurements have been made of the reactions of  $\text{NO}^+ (\text{a}^3\Sigma^+)$  ions:



(Glosik et al. 1978, Dotan et al. 1979.) The rate coefficients are  $k_{33} + k_{34} = 3.2 \pm 1.3 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  and  $k_{35} = 5.0 \pm 2.0 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ .

### 2.7. $N^+$ ions

During the period of this contract, we have studied the  $N^+$  composition in detail. The ions are removed by the reactions



In the analysis of the AE data we adopted the laboratory values (Smith et al. 1978) of  $k_{36} = 2.6 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ ,  $k_{37} = 3.1 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  and  $k_{38} = 3.7 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$ . At higher altitudes,  $N^+$  is also removed by



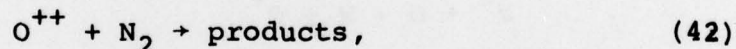
Our analysis of AE  $N^+$  data, particularly those taken during AE-C orbits 594 U and 2743 D leads to  $k_{39} = 2.2 \times 10^{-12} \text{ cm}^3 \text{ s}^{-1}$  and  $k_{40} = 3.6 \times 10^{-12} \text{ cm}^3 \text{ s}^{-1}$ . The probable uncertainty in  $k_{39}$  is 20% and of  $k_{40}$  30%. The value of  $k_{40}$  is in harmony with that derived from an analysis of observations of the planetary nebula NGC 7027 (Pequignot et al. 1978) and with a priori theoretical calculations (Butler and Dalgarno 1979).

The atmospheric temperature at which  $k_{39}$  was derived was about 840 K. Neynaber et al. (1977) have carried out preliminary measurements on reaction (39) which yield a cross section between  $2 \times 10^{-17} \text{ cm}^2$  and  $5 \times 10^{-18} \text{ cm}^2$  at energies between 5.0 and 0.5 eV. These measurements are at least not in contradiction to our derived rate coefficients.

### 2.8 Doubly-charged $\text{O}^{++}$ ions

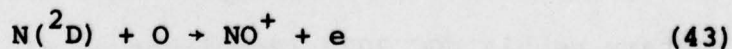


Breig et al. (1977) have analyzed the AE data on  $\text{O}^{++}$  and have concluded that  $k_{41} = 1 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$ . The analysis ignores the reaction



recently measured by Johnsen and Biondi (1978), and neglects a potentially large source of  $\text{O}^{++}$  (Victor 1979). A more careful study is needed before the empirical value derived from AE data can be accepted as plausible.

### 3. Associative ionization



It has been suggested by Zipf (1978) that reaction (43), which is endothermic by only 0.4 eV, may have important consequences on the derivations of the recombination coefficient of  $\text{NO}^+$  and on the conclusions about the deactivation of  $\text{N}(^2\text{D})$ . We have used the cross sections measured by Ringer (1978) to calculate the rate coefficients of reaction (43). The results are given in table 1. The reaction may be important at higher temperatures but is negligible in the normal atmosphere.

Table 1

Rate coefficient for  $N(^2D) + O \rightarrow NO^+ + e$

<u>T</u>	<u><math>k(\text{cm}^3\text{s}^{-1})</math></u>
500	$5.0 \times 10^{-17}$
1000	$4.6 \times 10^{-15}$
1500	$2.4 \times 10^{-14}$
2000	$6.0 \times 10^{-14}$
2500	$1.1 \times 10^{-13}$
3000	$1.8 \times 10^{-13}$
3500	$2.6 \times 10^{-13}$
4000	$3.5 \times 10^{-13}$
4500	$4.5 \times 10^{-13}$
5000	$5.7 \times 10^{-13}$

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