

The Photochemistry of Jupiter Above 1000 Å

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ABSTRACT

Jovian photochemistry above 1000 Å appears to consist of four zones: 1) photolysis of methane at 1216 Å and a total pressure of less than 10^{-5} atm; 2) photolysis of ammonia at 1700–2200 Å and a total pressure of less than 5×10^{-4} atm; 3) photolysis of both methane and ammonia at 1350–1450 Å at a total pressure of about 10^{-4} atm; and 4) photolysis of ammonia at 1450–1700 Å at a total pressure $< 10^{-4}$ atm. No photolysis occurs below this level. The large excess of hydrogen suggests that all radicals ultimately react with hydrogen, the net result being the production of H atoms. It is concluded that complex C–H–N molecules are not formed in Jovian photochemistry at wavelengths > 1000 Å because of the reducing atmosphere. Methane and ammonia persist in the atmosphere because photochemical mechanisms for their destruction ultimately results in their reformation by reaction of such species as CH_2 , NH and NH_2 with H_2 .

1. Introduction

Jupiter, whose mass is 1.8×10^{27} kg, or 317 times that of earth, consists of a dense central region of hydrogen about 7.1×10^4 km in radius (Wildt, 1961). The surface pressure¹ has been estimated to be at least 3×10^8 newtons m^{-2} (3000 atm) (Opik, 1962).

Moving outward from the central region the temperature falls and a dense cloud of solid ammonia is encountered which extends upward to a height of 50–60 km. At the cloud top where the pressure is about 1.1×10^6 N m^{-2} (11 atm) (Opik, 1962) [Another estimate is 2×10^5 N m^{-2} or 2 atm (Thornton and Welch, 1963).], the temperature is about 156K (Opik, 1962). Beyond the cloud top the temperature again falls and at a distance of about 120 km above the top (the stratosphere) it reaches about 80K (Trafton, 1964) as determined by estimating the scale height from the horizontal refraction during occultation of the light from the star α Arietis (Baum and Code, 1953). The total mass of the atmosphere is quite negligible compared with that of the condensed core. The problem of the temperature and composition of the Jovian atmosphere as a function of altitude is one of great complexity. The figures given here are subject to considerable uncertainty but even so are adequate to give a picture of the conditions under which the photochemistry occurs.

The solar spectrum to which planetary atmospheres are subjected has been measured (Detwiler *et al.*, 1961) from 850 to 2600 Å. With regard to the photochemistry of planetary atmospheres the most important set of information presented is the integrated intensity over 50 Å intervals which includes both continuum and lines. These data are set forth in Table 1. It is evident that radiation above 950 Å is much more prominent than that below this threshold to which Saslaw and Wildey

(1967) ascribe importance in Jovian photochemistry. While contributions to the photochemistry, particularly photoionization and subsequent ion-molecule reactions, are certainly made below 950 Å, photolysis above this threshold is much more prominent. It is apparent from Table 1 that between 2200 Å, where ammonia first begins to absorb strongly, and 1450 Å, where methane begins to absorb, the integrated intensity is about 100 times greater than that from 1450 to 850 Å.

From the standpoint of Jovian photochemistry, the only region of the atmosphere directly involved in photon absorption is the region above the cloud layer. That this is true is obvious from the composition of the atmosphere suggested by Opik (1962), Lasker (1963) and Trafton (1967). These atmospheric compositions are presented in Table 2. Since Trafton took account of both convective and radiative transfer in deducing the atmospheric composition, his model atmosphere

TABLE 1. Integrated intensity of solar spectrum in 50 Å intervals above the earth's atmosphere.

Center of interval (Å)	<i>I</i> (ergs $\text{cm}^{-2} \text{sec}^{-1}$)	Center of interval (Å)	<i>I</i> (ergs $\text{cm}^{-2} \text{sec}^{-1}$)	Center of interval (Å)	<i>I</i> (ergs $\text{cm}^{-2} \text{sec}^{-1}$)
2600	700	2050	90	1500	0.95
2550	560	2000	70	1450	0.50
2500	380	1950	55	1400	0.26
2450	390	1900	41	1350	0.26
2400	340	1850	28	1300	0.18
2350	320	1800	19	1250	0.15
2300	360	1750	12	1200*	5.7
2250	350	1700	8.2	1150	0.08
				1100	0.06
2200	310	1650	5.0	1050	0.10
				1000	0.18
2150	240	1600	3.2	950	0.15
				900	0.25
2100	145	1550	1.7	850	0.11

* Includes Lyman- α .

¹ 1 atm = 101, 325 N m^{-2} .

TABLE 2. Percentage composition of Jupiter's atmosphere according to various models.*

	He	H ₂	Ne	CH ₄	Ar	NH ₃ *
Opik (1962)	97.2	2.3	0.39	0.063	0.042	0.0029
Lasker (1963)	22.7	72.9	0.020	0.020	...	0.003
Trafton (1967)	66.6	33.3	...	0.10	...	0.0067

* Ammonia is thermodynamically very stable even at a pressure of 10⁻⁸ atm and if it were in equilibrium with its elements, almost no nitrogen would be present.

will be used in the calculations that follow. A cloud top pressure of 11 atm will be employed (Opik, 1962).

The average ratio of the absorption coefficients $\epsilon_{\text{NH}_3}/\epsilon_{\text{CH}_4} > 10^3$ from 1450–2200Å and the relative amounts I of this radiation absorbed by ammonia and methane (Watanabe *et al.*, 1953; Watanabe 1964) is given by the ratio $I_{\text{NH}_3}/I_{\text{CH}_4} > 10^3/15.0$. Thus, at least 98% of the absorbed radiation in the range 1450–2200Å is absorbed by ammonia. Further, if ϵ_{NH_3} is taken as 10 it may be seen that the topmost 0.01% of the atmosphere absorbs 99% of the incident radiation between 1450–2200 Å. The photochemistry in this region is essentially the photolysis of ammonia in the presence of a 15-fold excess of methane, 5000-fold excess of hydrogen and a 2500-fold excess of helium (Table 2). Moreover, about 99% of the solar photons capable of being absorbed by NH₃ and CH₄ are in the 1450–2200Å region (Table 1). The attenuation of light intensity normal to the surface of Jupiter as its atmosphere is penetrated is a function of the wavelength and may be approximated in the following crude way.

2. The region from 1450–1700Å

From 1450–1700Å only ammonia is capable of absorbing light and the average absorption coefficient² ϵ has a value of 10 atm⁻¹ cm⁻¹ ($\sigma = 3.72 \times 10^{-19}$ cm² molecule⁻¹) at 300K. The rate of attenuation of intensity with penetration is given by

$$dI/d(\infty - X) = -\epsilon PI = -\sigma nI, \tag{1}$$

where X is the distance above the cloud top, P the pressure of ammonia and n the number of molecules cm⁻³. An equivalent expression is

$$dI/I = \epsilon P dX = \sigma n dX. \tag{2}$$

The pressure falls with the increasing altitude X according to

$$P/P_0 = \exp(-X/H) = n/n_0, \tag{3}$$

where H is the scale height. The temperature is assumed to be constant and the atmosphere is assumed to be homogeneous. While these assumptions are undoubtedly incorrect (Greenspan and Owen, 1967), they are valid

² All absorption coefficients are given for 300K even though the actual temperature is much lower. The uncertainty in the pressures and composition of the atmosphere is too large to warrant correction of ϵ to the actual temperature.

TABLE 3. Attenuation of intensity by ammonia in the region 1450–1700Å as a function of distance X above cloud top.

I/I_0	X^*
0.995	14H
0.97	12H
0.77	10H
0.49	9H
0.14	8H
0.005	7H

* H is the scale height.

for the purpose of the present argument. The scale height is given by

$$H = kT/(\mu M_0 g), \tag{4}$$

where k is Boltzman's constant, μ the mean molecular weight of the atmosphere, M_0 the mass of unit molecular weight and g the gravitational acceleration. Then

$$dI/I = \epsilon P_0 \exp(-X/H) dX = \sigma n_0 \exp(-X/H) dX. \tag{5}$$

Upon integration, (5) becomes

$$I/I_0 = \exp[-H \epsilon P_0 \exp(-X/H)] = \exp[\sigma n_0 H \exp(-X/H)], \tag{6}$$

where $\sigma n_0 H = \tau$, the optical depth. Using this definition, (6) becomes

$$I/I_0 = \exp[-\tau \exp(-X/H)]. \tag{7}$$

According to Opik (1962), the cloud top, where the total pressure is 11 atm, is 6H above the dense inner region, and the pressure of ammonia P_0 at the cloud top is 70 N m⁻² (7×10^{-4} atm). The scale height H is 8.3 km and $\epsilon H P_0 = 5810$. Table 3 gives the calculated intensities as a function of X , the height above the cloud top and the corresponding partial pressures of NH₃. The number n (molecules cm⁻³) as a function of altitude is given by

$$n/n_0 = \exp(-X/H), \tag{8}$$

and the total number from $X=0$ to X by

$$\sum_0^X = \int_0^X n dX = \int_0^X n_0 \exp(-X/H) dX, \tag{9}$$

where n_0 is the number per unit height in a 1 cm² column at $X=0$. Integrating, we have

$$\sum_0^X = n_0 H [1 - \exp(-X/H)]. \tag{10}$$

When

$$X \rightarrow \infty, \quad \sum_0^X = n_0 H,$$

i.e., the total number of molecules in a 1 cm² column is given by the product of the surface (cloud top) concentration and the scale height. The fraction of the total

TABLE 4. Attenuation of intensity by ammonia in the region 1700–2200Å

I/I_0	X^*
0.95	14H
0.70	12H
0.40	11H
0.07	10H

* H is the scale height.

ammonia that lies below X is

$$\sum_0^X / \sum_0^\infty = 1 - \exp(-X/H), \quad (11)$$

and the fraction above X is

$$\left(\sum_0^\infty - \sum_0^X\right) / \sum_0^\infty = \exp(-X/H). \quad (12)$$

Since 99.5% of the radiation is absorbed above $X=7H$, it is evident that 99.5% of the radiation (1450–1700Å) is absorbed in the uppermost e^{-7} or 0.0009 of the ammonia where the maximum pressure is about 10^{-2} atm.

3. The spectral region above 1700Å

The sun's spectrum is most intense (Table 1) above 1700Å where the absorption coefficient of ammonia is about $100 \text{ atm}^{-1} \text{ cm}^{-1}$ and the photochemistry of ammonia is occurring at higher altitudes; Eq. (6) applies with a value of $H\epsilon P_0 = 5.8 \times 10^4$. The attenuation of intensity is presented in Table 4. It is evident that some 93% of the radiation above 1700Å is absorbed by the uppermost 0.000045 of the ammonia where the maximum total pressure is 5×10^{-4} atm.

4. The spectral region from 1000–1450Å

In the region below 1450Å, the most important line in the solar spectrum is Lyman α (1216Å) whose intensity is about equal to the total of all the rest of the integrated intensity from 1450Å to the x-ray region. The absorption coefficient of methane at 1216Å is about 500 but in the whole region the average is taken as about $300 \text{ atm}^{-1} \text{ cm}^{-1}$.

As before, we make use of Eq. (6), where the pressure of methane, according to Trafton (1967), is 15 times that of ammonia at the cloud top. Since the absorption coefficient of ammonia in the neighborhood of Lyman alpha is only about $190 \text{ atm}^{-1} \text{ cm}^{-1}$, absorption must be almost exclusively by methane. Thus, $P_0(\text{CH}_4) = 15 \times 7 \times 10^{-4} = 1.05 \times 10^{-2}$ atm, and using $\epsilon = 300 \text{ atm}^{-1} \text{ cm}^{-1}$ and $H = 8.3 \times 10^5$ cm, we have $\epsilon H P_0 = 2.61 \times 10^6$. The attenuation of intensity in the region 1000–1450Å by methane is given in Table 5. The fraction of methane lying above $13H$ (which absorbs 99.7% of the radiation between 1000–1450Å) is given by (12) with $X = 13H$, with the maximum total pressure being

TABLE 5. Attenuation of intensity by methane in the region 1000–1450Å.

I/I_0	X^*
0.995	20H
0.96	18H
0.75	16H
0.45	15H
0.12	14H
0.003	13H

* H is the scale height.

about 10^{-5} atm; thus,

$$\left(\sum_0^\infty - \sum_0^{13H}\right) / \sum_0^\infty = \exp(-13) = 2.25 \times 10^{-6}. \quad (13)$$

In the region 1000–1450Å some 85% of the radiation is the hydrogen atomic resonance line at 1216Å to which most of the photochemistry may be attributed.

In the spectral region 1350–1450Å (actually a very low solar intensity region) the absorption ratios are comparable as shown in Table 6. In this spectral region both methane and ammonia are absorbing appreciably. The height above the cloud layer at which this condition holds may be determined by adopting $\epsilon \sim 10$ for CH_4 . Then $\epsilon H P_0 = 10 \times 8.3 \times 10^5 \times 1.05 \times 10^{-2} = 8.72 \times 10^4$. If we set the absorption equal to 50%,

$$I/I_0 = 0.50 = \exp[-8.72 \times 10^4 \exp(-X/H)], \quad (14)$$

$$\exp(-X/H) = 7.94 \times 10^{-6}, \quad (15)$$

$$X/H = 11.7. \quad (16)$$

Thus, in the vicinity of 11.7 scale heights above the cloud top where the total pressure is about 10^{-4} atm, both methane and ammonia are being photolyzed (1350–1450Å).

5. Overview of primary processes

The overall photochemical picture above 1000Å is of four regions in the high Jovian atmosphere.

1) The uppermost region involves the photolysis of methane at (mostly) 1216Å in the presence of the other atmospheric constituents at a total pressure of about 10^{-5} atm. This region extends from 20 scale heights down to about 13 scale heights above the cloud top. In the region 10–14 scale heights, ammonia photolysis ($> 1700\text{Å}$) is also occurring.

TABLE 6. Relative absorption by methane and ammonia at wavelengths from 1350–1475Å.

$\epsilon_{\text{CH}_4} / \epsilon_{\text{NH}_3}$	λ (Å)
20	1350
0.5	1375
2.0	1400
0.3	1425
0.5	1450
< 0.1	1475

2) Around 11.7 scale heights both methane and ammonia are being photodissociated by very low intensity radiation in the 1350–1450Å region.

3) Below 13 scale heights and down to 7 scale heights ammonia is being photolyzed in the region 1450–1700Å.

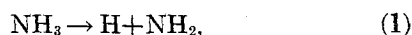
4) From 10–14 scale heights there occurs the photolysis of ammonia at wavelengths from 1700–2200Å and of extremely high intensity.

5) At altitudes less than 7 scale heights above the cloud top, virtually no photons above 1000Å are absorbed by gas phase molecules.

It is obvious that path lengths of many kilometers are required for simulation of Jovian atmospheric conditions if appreciable absorption is to be obtained for laboratory determination of the possibility of organic synthesis. Such simulated experiments have, of course, never been done. While Sagan (1961) attempted simulation experiments, whether the inevitable presence of the reaction vessel containing-wall influenced his results is a matter of considerable uncertainty.

6. Photochemical reactions

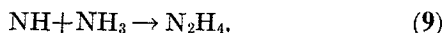
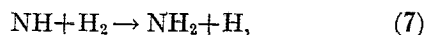
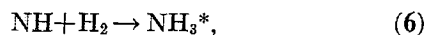
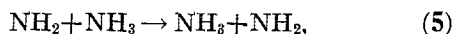
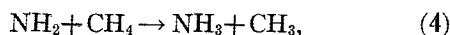
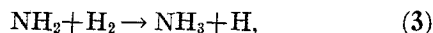
It has been shown (Bayes *et al.*, 1962) that the most important primary process in ammonia photolysis above 1600Å is the production of NH₂ according to



while from 1500–1600Å, NH is produced from



Because H₂ is several hundred times more abundant than methane which itself is 15 times more abundant than ammonia, the likely reaction partner of H, NH₂ and NH is H₂. Rate constants for the reactions of NH₂ and NH with H₂, CH₄ and NH₃ are not known but the reactions to be expected are as follows:

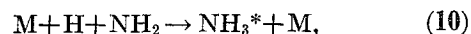


The following observations may be made: (i) reaction (5) is not a sink for NH₂; (ii) if the rate constants k_3 and k_4 are comparable, reaction (3) dominates reaction (4); and (iii) if the rate constants k_6 , k_7 and k_9 are comparable, reactions (6) and (7) dominate.

It is evident that reaction (1) followed by (3) constitutes a circuitous dissociation of H₂ with no net consumption of ammonia. Presumably, whatever hydrogen atoms are formed in these reactions ultimately

recombine with each other to form molecular hydrogen and the final result is simply a transfer of the photon energy to the Jovian atmosphere in the form of heat. The fact that no ammonia is consumed in this model accounts for the continued presence of ammonia in the atmosphere.

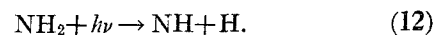
In considering the primary process (1) it is important to recognize that the energy of a photon of $\lambda > 1700\text{\AA}$ is insufficient to rupture a second N-H bond in addition to the first. The most abundant species present as a reaction partner of NH₂ is H₂. The kinetics of this reaction have not been measured but it is likely that an appreciable activation energy (~ 10 kcal) exists, i.e., the reaction is slow. Further, association reactions, e.g.,



are likely to be very slow in the region of pressure 10^{-4} – 10^{-5} atm. Thus, it is likely that NH₂ would have a relatively high photostationary state concentration were it not for the fact that it absorbs in the intense visible region from 4300–9000Å (Herzberg, 1966). The energy in this region is insufficient to dissociate the NH₂(A²A₁) state but the reaction of NH₂(A²A₁) with H₂ previously rejected for the ground state NH₂ may now proceed as shown in (11) if the radiative lifetime is at least comparable with the time between collisions with a hydrogen molecule, i.e., about 10^{-4} sec (or 10^{-6} sec if the Lasker atmosphere is assumed):



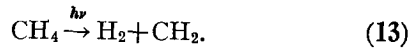
Alternatively, NH₂ may absorb dissociatively below 2800Å by



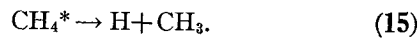
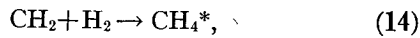
The resulting NH may then react with H₂ to reform ammonia according to reaction (6). However, the solar spectrum is vastly more intense in the green-red region than in the ultraviolet and NH₂ must primarily be pumped to the A state. Because of the abundance of helium, the A state may be relaxed by collision before reacting or radiating. It is to be expected, therefore, that if the A state radiates, the emission will be shifted to the red end of the usual red-green transition.

Because of the great prominence of Lyman α (1216Å) in the solar spectrum and its nearly exclusive absorption by CH₄, it is to be expected that the photochemistry above 14H will be controlled by methane. Here we exclude absorption by H atoms in Jupiter's atmosphere. The solar Lyman α is known to be strongly reversed (Ratcliffe, 1960) while the hydrogen atom absorption should be very sharp. However, the Doppler shift for the Jovian H atoms due to rotation (Carrington, private communication) of the planet is about 0.03Å and some solar Lyman α may still be absorbed by Jovian H atoms. It is unfortunate that the primary processes in methane photolysis are not very well understood. However, the most important process must be

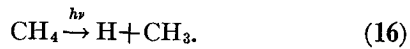
molecular elimination of H_2 (Mahan and Mandal, 1962) according to



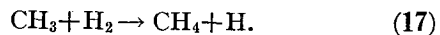
The CH_2 molecule reacts rapidly with the excess of H_2 to form an excited CH_4 which then decomposes (Bell and Kistiakowsky, 1962) to give



This sequence is certainly accompanied by an important primary process (Gordon and Ausloos, 1967; Laufer and McNesby, 1968)

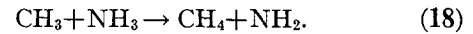


We are left with a free radical (CH_3) which, if it abstracts atomic hydrogen from H_2 , reforms methane by the reaction

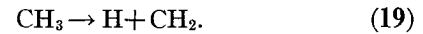


The result of the photolysis is, as in the case of NH_2 , a circuitous dissociation of H_2 . Of course, abstraction of H from CH_4 by CH_3 accomplishes nothing chemically. If CH_3 abstracts from NH_3 , methane is reformed and the whole process corresponds [reactions (16) and (18)]

to the dissociation of ammonia into H and NH_2 , i.e.,



As pointed out by Cadle (1961), reactions (17) and (18) have high activation energies and are improbable for ground state methyl radicals. However, reactions (17) and (18) may be rapid if the methyl radical becomes excited by absorption of longer wavelength sunlight. The possibility of the photolysis of CH_3 exists and this may produce CH_2 according to



If this occurs, the sequences (14), (15) will certainly follow. Cadle's argument concluded that methane and ammonia should have disappeared from the Jovian atmosphere because he rejected reaction (17) on grounds of too high an activation energy.

The generalization deduced from these arguments is that photolysis of a low pressure gas containing a large excess of H_2 , where much smaller amounts of CH_4 and NH_3 are the absorbers, can only result in heat being delivered to the atmosphere with no net chemical change. In this sense the chemical dynamics of the Jovian atmosphere are similar to those of the earth's where no net chemical change occurs in the O_3 , O_2 and N_2 system.

The suggestion (Sagan, 1961) that complex organic molecules may result from Jovian photochemistry

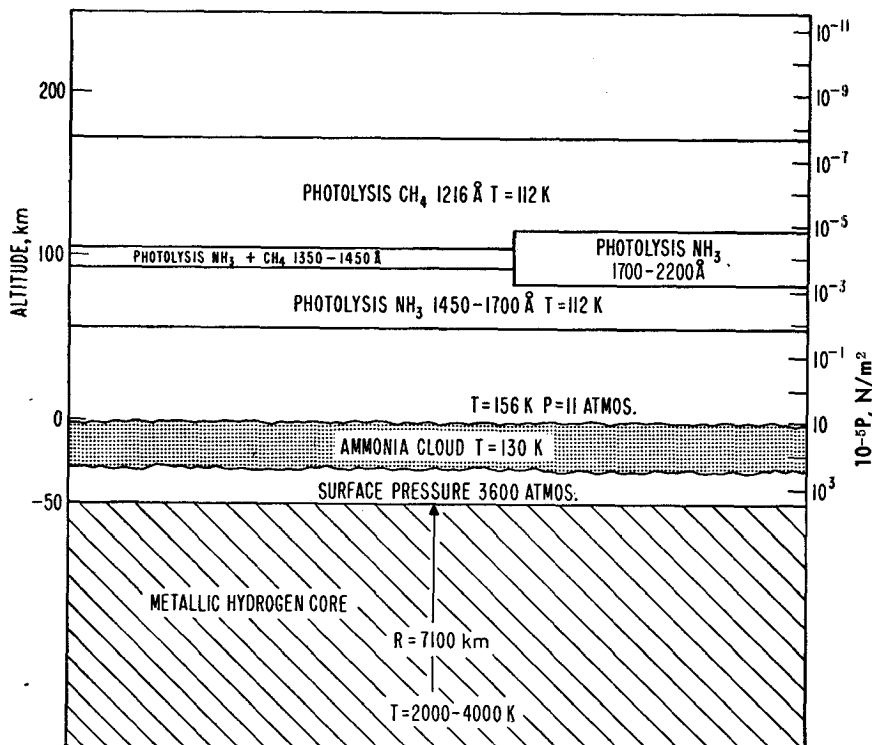


FIG. 1. Cross section of principal absorbing layers in the Jovian atmosphere.

because methane and ammonia are the absorbing atmospheric constituents is, therefore, doubtful, at least at wavelengths $> 1000\text{\AA}$. There is, however, some evidence that the passage of electrical discharges through simulated Jovian atmospheres does produce complex molecules (Sagan *et al.*, 1967; Miller, 1955). The extent to which these "simulated" atmospheres are perturbed by surface reactions is a matter of conjecture. Nevertheless, the formation of complex molecules by electrical disturbances in the Jovian atmosphere is certainly preferred to their formation by photolysis above 1000\AA . The problem of whether radiation below 1000\AA can produce complex organic molecules is a separate question which will not be discussed here.

Fig. 1 is a cross section of the principal absorbing layers in the Jovian atmosphere as discussed in this paper.

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