A theoretical study of the changes occurring in the ozonosphere during a total eclipse of the sun

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ABSTRACT

A theoretical investigation has been made into the variation of the ozone concentration in the atmosphere which results from an eclipse of the sun. The variation of the ozone concentration with time was followed by numerically integrating the relevant differential equations, allowing for the change in the solar radiation intensity reaching the earth's atmosphere during the eclipse. The results show that an increase of only 0.6 % occurs in the total ozone amount, and that the eclipse does not affect the ozonosphere below 45 km. The theoretical results are compared with experimental observations and reasons are advanced for the discrepancy between the two, which indicate that better agreement should be obtained when corrections for limb darkening of the sun are incorporated in the analysis of the experimental results.

1. Introduction

It is well known that an eclipse of the sun results in marked changes in the state of the upper atmosphere, particularly in the ionosphere where the changes can be studied easily by means of radio waves. At lower heights a solar eclipse also affects the ozonosphere, where it is found that an increase occurs in the total ozone content in a vertical column of the atmosphere. There have been several reported studies of this effect, the most recent and best documented of which are the observations of STRANZ (1961). Stranz, working from the Belgian Congo latitude 2° S, measured an increase of about 4% in the total ozone amount shortly after the maximum phase of an eclipse, in which only about 80% of totality was reached. Qualitatively it can be seen, from a knowledge of the diurnal photochemical atmospheric ozone variation and a study of the equations controlling the ozone concentration in the atmosphere, that an increase in the ozone concentration should occur above about 40 km as a result of a solar eclipse due to photochemical effects. However only about 1 % of the total ozone amount is present above 40 km in the atmosphere, hence Stranz's experimental results indicate that a four-fold increase takes place in the total ozone content above 40 km during a solar eclipse. Such an increase can be easily shown to be physically impossible and the question therefore arises as to what actually happens to the ozone concentration during an eclipse, and what is the cause of the apparent ozone increase observed experimentally. For these reasons it was thought that a theoretical study of the photochemical ozone variation during a solar eclipse might help to resolve these paradoxes, especially as no such study appears to have been made previously. The present investigation shows that theoretically an ozone increase of only about 1% is to be expected during a solar eclipse, and reasons are advanced to explain the difference between theory and experiment.

2. Reaction scheme

The differential equations governing the photochemical ozone and atomic oxygen concentrations were derived on the assumption that the only reactive species in the atmosphere was oxygen. This assumption is normally made since it permits the number of reactions which have to be considered to be reduced to five, and it also appears to give realistic results for

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the data used in this investigation. However the numerical values obtained for an oxygen atmosphere are likely to represent an upper limit to any variations which occur in the atmosphere during an eclipse, and this should be borne in mind when considering the results presented later.

The well known photochemical O_3 reaction scheme considered consisted of the following reactions:

$$O_2 + h\nu \longrightarrow O + O \quad \text{for} \quad \lambda < 2\,424\,\text{Å}, \quad (1)$$

$$O_3 + h\nu \longrightarrow O + O_2 \quad \text{for} \quad \lambda < 11\,800\,\text{Å}, \quad (2)$$

$$O + O_2 + M \xrightarrow{k_3} O_3 + M, \quad (3)$$

$$O + O_3 \xrightarrow{k_4} O_2 + O_2, \quad (4)$$

$$O + O + M \xrightarrow{k_5} O_2 + M. \quad (5)$$

From these reactions it follows that the differential equations governing the time variation of the O_3 and O concentrations are:

$$\frac{d[O_3]}{dt} = k_3[O][O_2][M] - \alpha_3 q_3 [O_3] - k_4[O][O_3], \quad (6)$$

$$\frac{d[O]}{dt} = 2\alpha_2 q_2[O_2] + \alpha_3 q_3[O_3] - k_3[O][O_2][M] - k_4[O][O_3] - 2k_5[O]^2[M], \quad (7)$$

where α_2, q_2 and α_3, q_3 are the absorption coefficients and the number of quanta of solar radiation absorbed by O_2 and O_3 respectively, and the bracketed terms refer to the concentrations of the various gases. M is any third body, normally nitrogen in the atmosphere.

The values of the absorption coefficients of O_3 were selected from the measurements of Tanaka *et al.* (1953), Inn & Tanaka (1953) and Vigroux (1952), the corresponding values for O_2 were taken from Watanabe *et al.* (1953) and Ditchburn & Young (1962). For the pressure dependent α_2 values of the Herzberg continuum the "vacuum" values of Ditchburn and Young were used because of the low pressures for the altitude range considered. The solar radiation intensity data selected were those given by Detwiler *et al.* (1961) and Johnson (1954). All of the above data have been tabulated in a previous equilibrium study of the photochemistry of O_3 (Hunt, 1963).

The following values were used for the rate constants.

$$k_3 = 3.7 \times 10^{-34} e^{-800/RT} \text{ cm}^6 \text{ molecules}^{-2} \text{ sec}^{-1}$$

from Zaslowsky (1962),

$$k_4 = 6.5 \times 10^{-12} e^{-3200/RT} \text{ cm}^3 \text{ molecule}^{-1} \text{ sec}^{-1}$$

from Leighton *et al.* (see Harteck & Reeves, 1961), and

$$k_5 = 2.7 \times 10^{-33} \text{ cm}^6 \text{ molecules}^{-2} \text{ sec}^{-1}$$

from Reeves *et al.* (1960).

The model atmosphere selected for this study was the 1959 ARDC atmosphere.

3. Numerical methods

3.1. General details

It is possible to study the photochemical variations of the O and O_3 concentrations with time in the atmosphere by numerically integrating equations (6) and (7) simultaneously, a procedure used previously by Leovy (1964) and Hunt (1965) to investigate the diurnal variation of O_3. The numerical methods used here were identical to those developed by Hunt (1965) for the diurnal investigation, except that the radiation terms in the equations had to be modified to allow for the variation of the intensity of the solar radiation reaching the atmosphere because of the occultation of the sun by the moon during the eclipse. The initial values of the O and O_3 concentrations required for the integration routine were also taken from the diurnal study, and corresponded to the non-equilibrium values obtained at noon at the equator for an overhead sun.

The time of the eclipse from first contact to fourth contact was taken as 9000 seconds, and at the instant of totality at 4500 seconds it was assumed that no solar radiation reached the earth. This is not strictly true in practice because of the existence of the corona around the sun, but since the intensity of the corona at the sun's limb is only 10^{-6} of the intensity at the sun's centre its presence was completely ignored throughout the investigation. For convenience the eclipse was considered to start at noon and during its course allowance was made for the variation of the zenith angle of the sun. After the completion of the eclipse the integra-

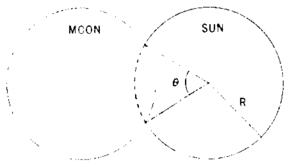

FIG. 1. The area representation of the sun during an eclipse.

tion was continued in order to determine how long the atmosphere required to achieve its normal O_3 and O concentrations. The time step for the integration routine was taken as 30 seconds, the same as that for the diurnal study, and was maintained constant with height, an empirical check being made to determine if truncation errors or other inaccuracies were present. The altitude range considered in the present investigation was limited to 48 to 80 km as in the previous diurnal study. The upper limit arises because of the rapid decrease in the O_3 concentration with altitude in the mesosphere, while the lower limit is determined by the reduction in the maximum permissible size of the integration time step at these lower levels. Also below about 50 km the present study is not of interest since the change in the O_3 concentration produced by the eclipse becomes negligibly small with decreasing altitude.

Two cases were considered in this study, a preliminary investigation in which no allowance was made for limb darkening of the sun, and the more realistic case in which allowance was made for limb darkening. Since the methods used to represent the variations in the solar intensity reaching the earth's atmosphere were basically different they will be discussed separately for convenience in the following sections.

3.2. The variation of the radiation term assuming no limb darkening

When the limb darkening of the sun is neglected the variation of the intensity of the sun's radiation reaching the earth is then directly proportional to the unocculted area of the sun, and can be defined in terms of the angle θ in Fig. 1.

Taking the apparent radii of the sun and moon equal to R we see that the occulted area, A_c, is given by

$$A_c = R^2[\theta - \sin\theta]$$

and if $$A_0 = \pi R^2$$

then $$A_\theta = A_0 - A_c = R^2[\pi - \theta + \sin\theta],$$

where A_θ is the unocculted area of the sun. If q_0 is the radiation output of the sun corresponding to the area A_0 then the radiation output q_0 for the unocculted area A_θ is

$$q_\theta = q_0 \frac{A_\theta}{A_0},$$

$$q_\theta = q_0 \left[\frac{\pi - \theta + \sin\theta}{\pi}\right]. \qquad (8)$$

Hence, by evaluating the coefficient of q_0 in (8), the effective radiation output of the sun can be determined for any value of θ.

3.3. The variation of the radiation term allowing for limb darkening

Limb darkening of the sun results from the effective radiation temperature of the sun decreasing towards the limb where the radiation is emitted from the cooler, outer layers. The radiation intensity for a given position on the sun is conventionally defined by the angle ψ as shown in Fig. 2. Hence we require to know the value of the radiation intensity $I(\psi)$ in order to allow for limb darkening. Values of $I(\psi)/I(0)$, where $I(0)$ is the intensity at the centre of the sun, have been given by MITCHELL (1959) and PEYTURAUX (1955) for $\cos\psi$ values from 0.2 to 1.0 and for various wavelengths down to about 3000 Å. Their data, which are plotted in Fig. 3, have been extrapolated as indicated since wavelength data were required to 1800 Å for the present study.

For a given wavelength PIERCE & WADDELL (1961) have shown that it is possible to represent the variation of $I(\psi)/I(0)$ with $\cos\psi$ by the following equation

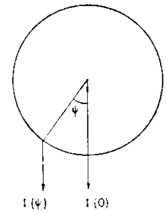

FIG. 2. The definition of a point on the sun's surface.

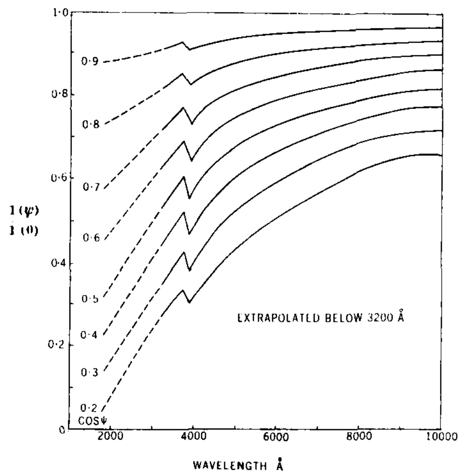

FIG. 3. The variation of $I(\psi)/I(0)$ with wavelength for different positions on the limb of the sun.

$$\frac{I(\psi)}{I(0)} = a_\lambda + b_\lambda \mu + c_\lambda[1 - \mu \ln(1 + \mu^{-1})],$$

where $\mu = \cos\psi$ and the coefficients a, b, c are functions of wavelength as indicated. The values of the coefficients a, b, c were determined for each of the 41 intervals used to represent the variation of the solar intensity with wavelength.

It is necessary to relate the value of $I(0)$ to that of F, the mean value of the intensity over the sun, as in the absence of any occultation of the sun the amount of radiation reaching the atmosphere for a given wavelength interval is given by F. The ratio $I(0)/F$ is plotted against wavelength in Fig. 4 from data given by PIERCE (1959), below 3200 Å it was again necessary to extrapolate the data as indicated. Because of the incorporation of limb darkening

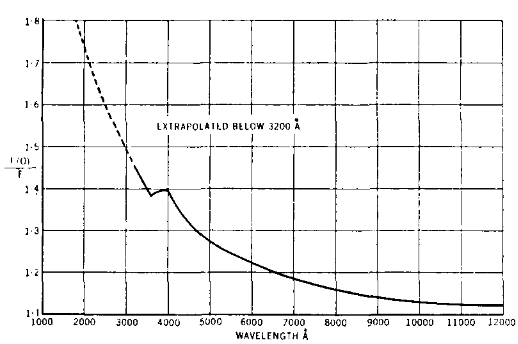

FIG. 4. Ratio of the intensity at the centre of the sun to the mean intensity for various wavelengths.

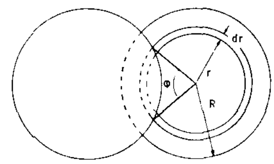

FIG. 5. The area representation of the sun allowing for limb darkening.

in the radiation terms in equations (6) and (7) the area representation given by equation (8) is no longer adequate and it is necessary to consider the surface area of the sun to consist of a number of annuli. For a given integration step the value of ϕ given in Fig. 5 was evaluated for each of the annuli which were occulted, the area of occultation being given by ϕrdr. The proportion of the mean value of the intensity, F, emitted by the occulted part of the annulus is then given by

$$I_c = \frac{\phi rdr.\, I(\psi)}{\pi R^2 F},$$

where $$\frac{I(\psi)}{F} = \frac{I(\psi)}{I(0)} \cdot \frac{I(0)}{F}.$$

The value of I_c, for each wavelength interval, was calculated for conditions corresponding to the middle of the integration time step. Then by summing over all the annuli occulted the reduction in the radiation intensity for the time step was obtained.

The resulting radiation intensity, corrected for atmospheric absorption, was then used with equations (6) and (7) to study the time variations of the O_3 and O concentrations during the eclipse. The numerical integrations were carried out on an IBM 7090 computer using a four point Runge-Kutta routine, which was found to give good accuracy and stability throughout the investigation.

4. Presentation and discussion of results

Only the results for the case in which limb darkening of the sun was considered will be presented here. The results were qualitatively very similar whether limb darkening was allowed for or neglected, but the magnitudes of the changes in the ozone concentrations were slightly greater for the limb darkening

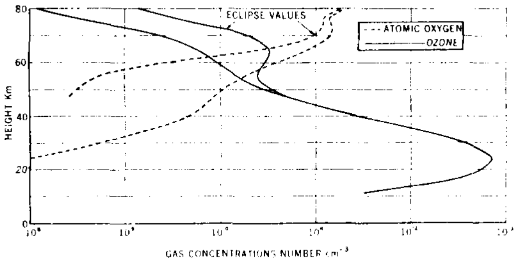

Fig. 6. *Comparison of the atomic oxygen and ozone* concentrations just after totality of the eclipse with their normal concentrations.

case. The reasons for the similarity of the results will be discussed later.

4.1. The variation in the atomic oxygen and ozone profiles due to the eclipse

The increase in the concentration of O_3 and the corresponding decrease in that of O due to the eclipse are indicated in Fig. 6, where the concentrations existing just after totality are compared with the "normal" concentrations for the same time of day. The profiles are plotted for the time just after totality at which the increase in the total O_3 amount is a maximum. Complete photochemical profiles are given in Fig. 6 in order to indicate more fully the extent of the changes occurring during the eclipse and their relative insignificance. There is a maximum increase of about one order of magnitude in the O_3 concentration above 70 km during the eclipse, but this increase falls off very rapidly with decreasing altitude, and below 45 km no change in the O_3 concentration can be expected. The variation of the O_3 concentration with time during the eclipse for several heights is shown in Fig. 7. The figure indicates the marked effect height has on the variation of the O_3 concentration and clearly shows the minor change which is obtained at 48 km. It should also be noted that the response of the ozonosphere to an eclipse varies with height, as might be expected, the maximum O_3 concentrations being obtained at different times for the various heights.

The increase in the O_3 concentration must obviously take place at the expense of the O concentration, and it can be seen from Fig. 6 that at all heights there is a reduction in its concentration which reaches over an order of magnitude at 60 km, and is even more accentuated at lower altitudes. Below 50 km the O concentration is much less than the O_3 concentration under normal conditions, hence even if all the O at these heights is converted to O_3 the percentage change in its concentration will be very small, and will rapidly approach zero with decreasing altitude. This is the reason why below 45 km the O_3 profile is not affected by the eclipse. At higher altitudes, above about 55 km, the concentration of O is greater than that of O_3 during the day, and relatively larger percentage changes can be expected in the O_3 concentration. However, at these heights the magnitude of the increase in the O_3 concentration is controlled by the losses caused by reaction (4) and not just by the rate of formation given by reaction (3). Above about 70 km the O concentration remains essentially unchanged by the eclipse in spite of the large O_3 increase, since a very small change in the O concentration at these levels can produce a large change in the O_3 concentration because of the two orders of magnitude difference in their relative concentrations. The reason an increase occurs in the O_3 concentration during an eclipse is of course due to the reduction in the intensity of the solar radiation reaching the atmosphere, which reduces the rate of dissociation of both O_2 and O_3 very markedly and allows the collision reactions to predominate as discussed above. In this respect the changes produced in the ozonosphere by an eclipse of the sun have many similarities to those produced by sunset where the same qualitative changes occur.

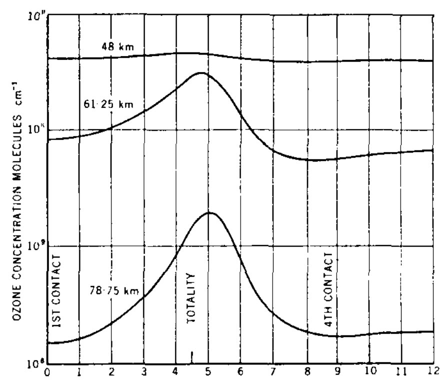

Fig. 7. The variation of the ozone concentration at various altitudes during an eclipse.

4.2. The variation of the total ozone amount during an eclipse

The theoretical increase in the total O_3 amount in a column of the atmosphere during an eclipse is perhaps the most interesting result of the present investigation, since experimental data are available for comparison. The calculated change in the total O_3 amount with time is given in Fig. 8, from which it can be seen that the maximum increase of 0.0016 cm STP is attained 4830 seconds after the start of the eclipse, or 330 seconds after totality. The maximum occurs after totality rather than at totality, since the radiation intensity reaching the atmosphere takes several minutes to reach any significant level after third contact, and during this time the collision reactions continue to predominate in the atmosphere. The rise in the O_3 content is very slow initially during the eclipse but increases as the percentage of the exposed area of the sun is reduced, after the maximum is attained there is a much more rapid decrease, the O_3 increase being reduced to zero in only 2000 seconds compared with a build up time of 4800 seconds. An interesting point is that the decrease in the O_3 content continues after the "normal" O_3 value for the corresponding time of day is reached, and a minimum occurs at about 8500 seconds, or 500 seconds before fourth contact, when there is a "negative" value of 0.0002 cm STP. The O_3 content then shows a small steady increase back to the "normal" value, the restoration time being greater than two hours.

If we take a total O_3 value of 0.27 cm STP as being representative of the O_3 content at the equator, then the maximum percentage increase in the total O_3 amount during an eclipse will be 0.6%, using the data given in Fig. 8. This is considered to be significantly different from the value of 4% reported by Stranz (1961) for a similar total O_3 amount. Eclipse observations made by Fournier d'Albe & Rasool (1956) at the experimentally difficult time near sunset failed to reveal any noticeable change in the total O_3 amount, and they obtained only irregular and minor variations. Two eclipse observations have also been reported by Bezberkhny *et al.* (1956) who appear to have recorded O_3 increases of between three and four times the total O_3 amount, which are of course physically impossible and must presumably have been due to instrumental error.

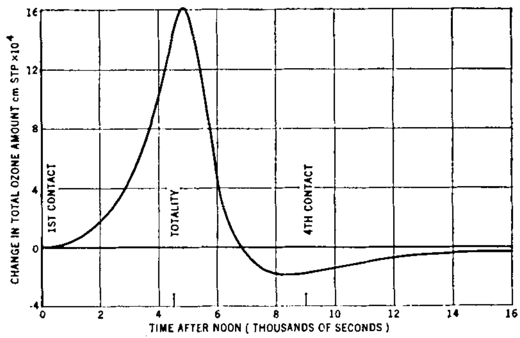

Fig. 8. The change in the total ozone amount in a column of the atmosphere during a solar eclipse.

4.3. The reason for the discrepancy between the theoretical and experimental results

The difference between the theoretical increase in the total O_3 amount and that observed experimentally is most likely to be due to inadequacies in the Dobson spectrophotometer, which is the standard instrument used in O_3 observations. This instrument is designed to measure the ratio of two wavelengths, one centred at about 3000 Å, the other at about 3300 Å. The total O_3 amount in the atmosphere is obtained from the measured ratio using the following formula:

$$\frac{I_1}{I_2} = \frac{I_{1,0}}{I_{2,0}} \exp\left[-(\alpha_1-\alpha_2)\mu X-(\beta_1+\beta_2)m\right], \quad (9)$$

where I_1/I_2 is the ratio of the intensities measured at the surface, $I_{1,0}/I_{2,0}$ is the ratio outside the earth's atmosphere, α is the O_3 absorption coefficient, β is a scattering coefficient, μ and m allow for the atmospheric path-length and X is the total ozone amount.

Now in order to calculate the total O_3 amount during an eclipse it is necessary to have a value of $I_{1,0}/I_{2,0}$, and it is normally assumed that this ratio remains constant and equal to its standard value, which is unlikely because of limb darkening of the sun. An estimate was made of the variation of this ratio during an eclipse and it was found that it fell to 97.8% of its initial value at 3600 seconds, and to 95.5% at 4200 seconds. From equation (9) it can be seen that any variation in this ratio will affect the apparent value of X, hence the incorporation of a limb darkening correction might be expected to reduce the value of X observed. It should be emphasized that if the theoretical results presented here are valid then no increase in the O_3 content of the atmosphere can be expected to be measured during an eclipse, because the theoretical increase of 0.6% is less than the quoted accuracy of 2% of the spectrophotometer.

Throughout this study the effect of the sun's corona has been neglected because of its low intensity, and although it is admitted that the corona would be important in determining the value of $I_{1,0}/I_{2,0}$ near totality, and therefore probably contributes to the discrepancy between theory and observation, its neglect is considered to be entirely justified despite the comments of BEZBERKHNY *et al.* (1956). They state that because it can be shown the equilibrium O_3 concentration depends on the ratio of the amount of radiation absorbed by O_2 to that absorbed by O_3, then the radiation near totality will be important because this ratio increases for the corona, although the absolute values are minute, indicating that an increase in the equilibrium O_3 amount is to be expected. This argument is erroneous as one is not dealing with an equilibrium situation but one in which the changes in the O_3 concentration depend on the magnitudes of the various terms in equation (6), so that absolute and not relative values are important.

4.4. THE EFFECT ON THE THEORETICAL RESULTS OF NEGLECTING LIMB DARKENING OF THE SUN

As stated previously neglecting limb darkening altered the results obtained quantitatively, but not qualitatively. Such a result is predictable since the predominant effect occurring during an eclipse is the overall reduction in the solar radiation intensity reaching the atmosphere, the relative change in this intensity with wavelength being of secondary importance. For about the first 3000 seconds of the eclipse the variation of the radiation intensity is essentially constant with wavelength, and is proportional to the exposed surface of the sun. Hence up to this time the neglect of limb darkening would be of minor importance, as the results were actually found to show. After this time the reduction in the exposed surface of the sun towards second contact progressively increases the importance of limb darkening, since the radiation becomes increasingly emitted from the limb itself. Fig. 3 shows that the effect of the limb darkening is most important for the shorter wavelengths, where the radiation intensity rapidly drops towards zero as the limb is approached. At a time of 3600 seconds the radiation intensity at 1800 Å has fallen to about 15% of its initial value, while at the long wavelength end of the scale, 7500 Å, the reduction is to only about 23% of its initial value. Now it is the shorter wavelengths, above about 3000 Å, which are principally absorbed at the upper levels of the ozonosphere, resulting in the dissociation of the O_3, hence the amount of O_3 dissociated becomes progressively less towards second contact compared with that when darkening is neglected. A larger increase in the total O_3 amount is therefore to be expected for the case in which limb darkening of the sun is allowed for. The final result of allowing for limb darkening was found to be of little importance as regards the increase in the total O_3 amount, a rise from 0.0013 to 0.0016 cm STP was observed due to limb darkening. However, the changes in the O_3 concentration above about 70 km were more marked. Nevertheless, it is apparent that no great error would have resulted if limb darkening had been neglected and the numerical study would have been greatly simplified.

5. Conclusions

The theoretical study made here of the changes occurring in the ozonosphere during a total eclipse of the sun has shown that an increase of less than 1% of the total O_3 amount is to be expected, compared with experimental observations of 4%. Correction of the experimental values to allow for limb darkening of the sun would give better agreement between theory and experiment. However it must be emphasized that the results presented here are for an oxygen atmosphere, and if allowance had been made for the presence of atomic hydrogen in the atmosphere the total O_3 increase should be even less than the value of 0.0016 cm predicted here. Indirect information regarding the changes in the O_3 concentration at various heights during an eclipse may be obtainable from airglow measurements of the emission of the hydroxyl bands and of the infrared atmospheric bands of oxygen.

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