

SODIUM IN THE UPPER ATMOSPHERE*

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ABSTRACT. The paper first surveys the available data for the sodium phenomena in the upper atmosphere, namely, intensity variations of the D-lines in the night air-glow and in the twilight flash, the height of the emitting layers, the distribution of sodium in the atmosphere and its probable source. The various excitation processes, both regarding the twilight flash and the night air-glow emission are closely examined. It is shown that the most probable mode of production of the twilight flash is resonance excitation of neutral sodium atoms in 35-65 km. region by solar radiation $\lambda 5893$, there being no significant effect of ozone screening. The possible excitation processes in the night air-glow are examined critically on the assumption that the height of the emitting layer is 250 km. (as obtained by Barbier and Roach) and that above 100 km. sodium exists wholly in the ionized state (as shown by Bates). Two possible excitation processes are examined: (1) Radiative recombination of Na^+ ions and (2) mutual neutralization of Na^+ and O^- ions. Of these two processes the latter seems more probable as the former requires impossibly high concentration of Na^+ ions. But the latter process cannot maintain the observed intensity of the radiation throughout the dark hours of the night. Hence it is concluded that extra terrestrial particles must be bombarding the upper atmospheric regions, ionizing and/or exciting the neutralized Na^+ ions. Alternatively, it may be assumed that sodium atoms are entering the atmosphere from interplanetary space.

I N T R O D U C T I O N

The presence of the element sodium in the upper atmospheric regions has been proved beyond doubt by the identification of the yellow D-lines in the night air glow spectrum. A large number of workers have made observations on the intensity variation of these lines both in the night air-glow and in the twilight flash, with a view to determine the height of the emitting layer, the distribution of the sodium content with height and also the total number of sodium atoms. Unfortunately, the results obtained are not concordant. For example, while according to some observers the sodium is concentrated in a comparatively thin layer between 70 and 110 km., others estimate the height of emission of the D-lines to be 250 km., *i.e.*, in the region of the F-layer of the ionosphere. Further, the modes of excitation of the sodium D-lines, both in the night air-glow and also during the twilight flash, are still insufficiently understood. The purpose of the present paper is, firstly, to give a connected account of our present state of knowledge of upper atmospheric sodium, and secondly, to examine and extend the theories of the excitation of the D-lines, and to deduce therefrom the probable distribution and source of sodium in the upper atmosphere.

* Communicated by Prof. S. K. Mitra

PRESENT STATE OF KNOWLEDGE OF UPPER
ATMOSPHERIC SODIUM

Sodium gives one of the most prominent and easily observed lines in the spectrum of the night air-glow. It was first identified by Bernard (1938) by accurate measurement of wavelength with Fabry-Perot interferometer.

Intensity and its variations.—The absolute intensity of the radiation has been measured by several authors (Cabannes, Dufay and Gauzit, 1938; Barbier and Roach, 1950a; Bates and Nicolet, 1950) and has been found to correspond to 2×10^8 to 8×10^7 transitions per sec. per cm^2 column of atmosphere.

The intensity of the D-emission remains fairly constant throughout the night (Elvey and Farnsworth, 1942). During the morning and evening twilights, however, the sodium light is enhanced in the illuminated upper atmosphere to 50 to 100 times the night air-glow intensity. This enhancement is generally referred to as the *twilight flash* of sodium.

The intensity is, however, subject to considerable fluctuations from night to night. There may be nights when under the most favourable conditions the lines are absent from the twilight spectrogram (Vegard and Tönsberg, 1941). Further, the emission is not uniform from all parts of the sky.

There is also a seasonal variation in the intensity with a maximum in winter and a minimum in summer.

Height of the emitting layer—Many attempts have been made to determine the height of the layer emitting the D-lines in the night air-glow by the well known van Rhijn technique. The results obtained, however, are widely divergent amongst themselves as will be seen from Table I.

TABLE I

Observer	Height obtained	Remarks
Cabannes, Dufay and Gauzit (1938)	130 km.	Based on observations by Garrigue at Pic-du-Midi, France in 1936.
Dufay and Tcheng Mao-Lin (1948)	80 km.	Used only two zenith distances.
Barbier and Roach (1950a, Roach and Petit, 1951).	250 km.	Used interference type filters to transmit narrow bands, in conjunction with a photo-electric photometer

Observations on the twilight flash.—Bernard (1938) was the first to make systematic observations on the twilight flash. He found that the flash disappeared when the edge of the shadow cast by the earth passed at a height of about 60 km.

Vegard and Tönsberg (1940; 1941) found that the upper limit to the height of the emitting layer as deduced from observations of the twilight flash made near the horizon always gave a much larger value than those deduced from similar observations made near the zenith. This discrepancy they explained by assuming that the excitation of the D-lines was caused by ultraviolet light in a wave-length range that is absorbed by ozone. When calculating the height of the upper limit they, therefore, took into account the effective increase in the radius of the earth shadow by the ozone screening. From a large number of observations made at Oslo and Tromsø Vegard and Tönsberg determined the value of the screening height and also the upper limit of the height at which the flash disappeared. The mean value of the former (screening height) was found to be 49 km. and of the latter 110 km.

Cario and Stille (1950) made similar experiments over northern Germany in 1941. They measured the upper limit of the height of emission to be 118 ± 2.5 km and the screening to be 54 ± 3 km.

Barbier (1948) from twilight observations in Haute-Provence (France) estimated the base of the sodium layer at 70 km.

In addition to the ordinary twilight flash, as described above, a late twilight effect consisting of a slow steady decrease of the intensity for an hour or so after the astronomical twilight has also been observed by Barbier and Roach (1950b). Identical observations have been recorded for both evening and morning twilights. The measurements indicated the region of emission in the range 200 to 600 km. (However, in a private communication Roach states that the late twilight flash was not observed again and that he is inclined to believe that it is a sporadic phenomenon).

Distribution of sodium in the upper atmosphere.—Attempts have been made to determine the location and distribution of sodium in the upper atmosphere from observations on the night air-glow emission as also from those on the twilight flash. The results obtained by the different workers are as follows:

An estimate of the total number of sodium atoms in the upper atmosphere has been made by Bates and Massey (1946) from the observed intensity of the twilight emission and the flux of solar quanta (taking account of Fraunhofer absorption), and using the known probability of transitions yielding the D-lines. The estimated number of sodium atoms above 70 km. was found to be of the order 10^9 per cm^2 column of atmosphere.

Barbier (1948) has also given a similar estimate from twilight observations, namely, that in the 70 to 100 km region the number of sodium atoms per cm^2 column is 5×10^8 . It will be seen that according to both the estimates the total sodium content is only a minute fraction (10^{-12}) of

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the total atmospheric constituents. If it is assumed that sodium in the above proportion is distributed over the whole atmosphere then the number per cm^2 column would be 2×10^{13} .

In view of its minute proportion, the interesting suggestion has been made by Bates (1950) that it can be increased by a significant amount by ejecting sodium vapour in, say, 70 km. region from a rocket. This would enable controlled experimental observation on sodium emissions to be carried out.

An estimate of the thickness of the sodium layer was made by Vegard and Tönsberg (1940; 1941) by observing the time at which the intensity of the twilight flash began to drop rapidly (as the sun sank more and more below the horizon) and the time at which it vanished *i.e.*, fell to the night-time value. The former gave the time at which the shadow formed by the ozone screening sphere passed the lower border and the latter the time at which it passed the upper limit of the sodium layer. The thickness of the layer was obtained from a knowledge of this time interval and the height of the ozone screening sphere (*vide supra*). The thickness was found to vary between 8.4 and 27 km (with a mean value of 16.2 km). Hence it was concluded that the sodium producing the twilight flash was situated in a comparatively thin layer between the heights 85 km. and 110 km.

Elvey and Farnsworth (1942) put a different interpretation to the phenomenon of sudden disappearance of the twilight flash. This, according to the authors, was not due to the edge of the shadow having passed above the sodium layer, but was merely an effect of the sodium content falling exponentially to a low value. According to the observations of these authors, the decrease of intensity with height followed a logarithmic law and ran parallel to the decrease of number density of atmospheric molecules with height.

Barbier and Roach (1950a; also Roach and Pettit, 1951) have estimated the height of the emitting layer (see Table 1) to be around 250 km. *i.e.*, in the F-region of the ionosphere. In view of the many precautions taken by these authors the results obtained appear to be more reliable than those obtained by previous workers. This observation together with those on the twilight flash leads one to conclude that sodium is not confined within a narrow layer (as had been supposed by some authors), but is distributed in two layers: one in the 80 or 85 to 110 km. region, and another in the region of the F-layer of the ionosphere. Or, it may be that sodium in the atmosphere extends from a level of about 80 km. upto the F-region. (See however, discussion in the next section. The base of the sodium layer is taken at 35 km. and not at 80 km.).

Source of the upper atmospheric sodium.—Nothing definite is known about the source of the upper atmospheric sodium. The sodium may be of terrestrial origin, but there are strong reasons to believe that at least a part of the sodium comes into the atmosphere from outer space.

In favour of the terrestrial origin it has been suggested that ascending air currents may carry sodium salt from ocean sprays. Volcanic dust containing sodium compounds have also been known to shoot up to great heights—30 km. (Bernard, 1939).

Cabannes, Dufay and Gauzit (1938) and others attribute the origin of the atmospheric sodium to meteorites. The D-lines have been detected in meteoric spectra. But, it has been argued that the lines may be due to excitation of *atmospheric* sodium atoms (Roach, 1949).

Bates (1947) favours a cosmic origin from consideration of the density gradient of atmospheric sodium obtained from twilight investigations. Vegard (1940) suggests that sodium comes into the atmosphere from the sun along with the solar corpuscular streams that produce auroral and magnetic disturbance phenomena.

It has also been suggested that the earth is sweeping through interplanetary sodium clouds and the sodium is swept into the earth's atmosphere as the latter moves through space (Barbier and Roach, 1950*b*). This hypothesis promises an explanation of the seasonal variation of intensity of nocturnal D-lines.

It is difficult to judge the relative merits of the different hypotheses because we do not know how and at what rate the sodium is disappearing from the regions from which it emits light.

EXCITATION PROCESSES

(a) *Twilight flash*.—There have been two views regarding excitation of the sodium atom in the twilight flash.

According to Vegard and Tönsberg (1940, 1941) the exciting radiation is in the ultraviolet lying between 1900Å to 2900Å. They arrive at this conclusion from the fact that according to their observations the exciting radiation has to pass above the ozone layer (*vide supra*). Also the exciting radiation cannot be less than 1900Å because such radiation will be absorbed by the overlying mass of air. The wavelength, also, cannot be longer than 2900Å as otherwise ozone screening (Hartley bands) will not have any effect.

According to Barbier and Roach (1950*b*), however, the twilight flash is simply a case of resonance excitation, the active wavelength being 5893Å. They take into account the effect of ozone screening, but this effect is assumed to be due to the feeble absorption in the region of Chappuis bands and not to the strong Hartley bands.

A close examination of the two hypotheses shows that neither is wholly correct. The twilight flash is due to resonance excitation as proposed by Barbier and Roach, but the exciting radiation (5893Å) need not pass above the ozone layer. This will be clear from the consideration of the maximum possible absorption which can be effected by ozone in the region of Chappuis bands. Barbier, Chalonge and Vigroux (1942) made observations during a

lunar eclipse when they followed spectroscopically the sunlight passing obliquely through the earth's atmosphere on the darkening moon. Using absorption in the region of Chappuis bands they determined the ozone masses for the different ray paths which passed over the earth's surface within 4 to 17 km. They found that the maximum value of the ozone mass traversed was 11 cm. and that this occurred when the distance of the ray from the earth's surface was 12 km. Since the maximum absorption coefficient in the Chappuis bands is 0.05 at 6100\AA , the maximum reduction in intensity of solar radiation may be 40%. If the solar ray passes by the top of the ozone layer (40 to 50 km.) the absorption will be insignificant. It is, therefore, difficult to see how the resonance excitation process of sodium atoms by $\lambda 5893$ can be significantly affected by the ozone absorption in the region of Chappuis bands.

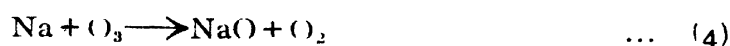
One has, however, to explain the observations of Vegard and Tönsberg namely, that there was a systematic difference between the height measurements of the upper limit of the sodium flash when observed along the zenith and when observed along a direction close to the horizon (and which was interpreted as due to the exciting radiation having to pass above the ozone layer). It should, however, be remembered that the results of height determinations, as carried out by Vegard and Tönsberg, were not sufficiently accurate. The spectrographs in the experiments were exposed for 3–6 minutes at the zenith and 6–10 minutes and sometimes 20–30 minutes at the low angles. If the time of disappearance occurs within the exposure interval it cannot be noted with accuracy; and an inaccuracy of 5 minutes would lead to an error in the height measurement of about 30 km. The conclusion of Vegard and Tönsberg, therefore, that the exciting radiation has to pass above the ozone screening height, is not fully warranted. It may, therefore, be concluded that the twilight flash is caused by resonance absorption of radiation $\lambda 5893$ and that any increase in the screening radius of the earth is only due to terrestrial absorption due to haze etc. extending up 5 km. at a liberal estimate.

A result of the above conclusion is that the height of the sodium layer as measured from observations of twilight flash by Vegard and Tönsberg, as also by Barbier will have to be reduced by about 45 km. This means that the bottom of the sodium layer, as indicated by the lower limit of the flash (corresponding to the time at which the intensity of the flash begins to drop rapidly), is at 35 to 40 km. and the top, as indicated by the upper limit, is at about 65 km. (of the same order as obtained by Bernard, who did not consider ozone screening). This result is more in conformity with the findings of Bates (1947), according to whom, due to the ionizing action of solar radiation, sodium above 80 or 90 Km. will almost wholly be in the ionized state. And, in such case, in the absence of neutral sodium atoms, no twilight enhancement is possible, though according to observations of Vegard and Tönsberg and Barbier twilight flash is observed (assuming ozone screening) even up to 110 km.

(b) *Nocturnal emission.*—To explain the nocturnal emission of the D-lines the presence of neutral sodium atoms is generally assumed. According to Chapman (1939) the energy of excitation of the sodium atom is derived from the solar energy which has been spent in dissociating the O_2 molecules in the upper atmosphere. Thus we may imagine the following reactions :



In reaction (1) the presence of the compound NaO is assumed. This may be produced by the following processes :



In reaction (3) M is the third body which carries away the excess of energy and momentum.

It may also be mentioned that Penndorf has sought to explain both the night emission and the twilight flash by a process in which Na_2O is produced through the presence of N_2O in the upper atmosphere (Penndorf, 1950). The region of emission is assumed to be 90 to 105 km.

Bates and Nicolet (1950) have made a close examination of the various processes that have so far been suggested and have come to the conclusion that for any of these processes to be effective, the effective level of emission cannot be much above 70 km.

Now, as already mentioned, the mean height of the emitting layer, according to measurements of Barbier and Roach is 250 km. In view of the considerable care which these authors have taken in their measurements, the result cannot be seriously in error. It, therefore, appears that none of the reactions proposed above can be responsible for the D-line emission. Further, it has been shown by Bates (1947) that at heights above 100 km. sodium is present only in the ionized state. One is thus forced to the conclusion that it is the Na^+ ions (and not neutral sodium atoms) that take part in the emission mechanism.

This conclusion raises the following problems regarding the D-line emission.

(i) What is the most likely process of neutralization of the Na^+ ions by which the Na atom produced is excited to the $2p$ level ?

(ii) What is the concentration of the Na^+ ions necessary in order that the radiation may have the observed intensity, namely, that corresponding to 8×10^7 transitions per cm^2 column ? How far is this concentration compatible with the observed ionization density in the F-region ?

(iii) Is the initial concentration of Na^+ sufficiently high to maintain the intensity of radiation throughout the dark hours of night with very little decrease as observed ?

Let us discuss these problems as far as possible with the available data and with the contemporary knowledge of the physical state of the upper atmosphere.

Emission processes (with Na^+ ions).—In view of the height of emission being 250 km., the region of emission may be identified with the F-layer of the ionosphere. This region contains N_2 molecules and O atoms, positive ions N_2^+ and O^+ , negative ion O^- and electrons. It also contains possibly N atoms.

We first consider the process of radiative electronic recombination,



The resulting sodium atom may be in the ground state or in any one of the excited states from which transitions may occur to the ground state (directly or in successive steps). Apparently, in this process lines of sodium other than $\lambda 5893$ would also be present. The presence of these lines has not yet been established, although detection of $\lambda 3303$ has been reported by some workers.

We next consider the mutual neutralization of O^- and Na^+ ,



The process may be expected to have a high probability when the resonance condition is satisfied, i.e., when the energy released on neutralization is taken up wholly as energy of excitation of the reaction products. Since the ionization potential of sodium is 5.1 eV and the electron affinity of oxygen is 3.0 eV (Vier and Mayer, 1944), the energy released on neutralization is $5.1 - 3.0 = 2.1$ eV. Since this is also the energy of excitation of the sodium atom to the $2p$ -state there is energy balance. Hence the mutual neutralization of Na^+ and O^- ions may be regarded as a very likely process for the emission of the D-lines.

Calculation of Na^+ ions necessary.—We now calculate the number density of Na^+ ions necessary to produce the observed intensity of radiation by the neutralization processes (5) and (6).

Let us consider first process (5). We assume that the height distribution of electrons follows a parabolic law in the region of maximum number density (as in a Chapman layer). The distribution of Na^+ ions is not known with any certainty. It may decrease exponentially following the decrease of atmospheric density, or there may be region of maximum concentration as in ionospheric layer formation. However, to simplify the calculation, we

will assume a constant concentration with height for the Na⁺ ions. We shall presently see that so far as qualitative result is concerned, this assumption does not lead to any erroneous conclusions.

With the above assumptions the number of transitions per cm³ at height h is given by

$$\alpha n^+ n_e(h)$$

where α is the coefficient of recombination and n^+ and $n_e(h)$ are the number densities of sodium ions and electrons respectively at the height concerned. Bates (1947) has given a value 2×10^{-12} cm³ per second for the total recombination coefficient i.e., for electrons captured in the ground state or in any of the possible excited states. But the electrons captured in the ground state or in any of the p -states, other than the $2p$ -states, will not contribute to the D-lines. Hence the value of the recombination coefficient for only the states that are of interest to us will be somewhat lower. We assume this value to be 1×10^{-12} cm³ per second.

Since we have assumed that the variation of electron concentration with height follows a parabolic law, we may write

$$n_e(h) = n_e(o) \left\{ 1 - \frac{h^2}{4H^2} \right\}$$

where $n_e(o)$ = number density of electrons in the region of maximum ionization = 5×10^5 per cm³.

H = scale height in the F-region at night = 50 km.

We assume that the bulk of the electrons in the parabolic layer lies within the heights $\pm 2H$, above and below the level of maximum concentration. Hence, since the number of transitions per cm³ per sec. at height h is $\alpha n^+ n_e(h)$, we obtain by integrating over the whole layer the number of transitions per sec. per cm² column,

$$\begin{aligned} & \alpha n^+ n_e(o) \int_{-2H}^{+2H} \left(1 - \frac{h^2}{4H^2} \right) dh \\ &= \alpha n^+ n_e(o) \times \frac{8}{3} H. \end{aligned}$$

Since this must be equal to the observed number of transitions, namely, 8×10^7 per sec. per cm² column we have for the concentration of Na⁺ ions,

$$\begin{aligned} n^+ &= \frac{8 \times 10^7 \times 3}{\alpha \times n_e(o) \times 8H} \\ &= 1.2 \times 10^7 \text{ per cm}^3. \end{aligned}$$

But this concentration is improbably high because the electrons produced by the ionization would raise the concentration of electrons also to the same order. We know, however, from radio measurements that the maximum concentration of electrons in the F-region is of the order 10^5 to 10^6 per cm³.

Calculations, exactly similar to the above, may also be carried out for process (6). The coefficient for the process may be assumed to be of the order 10^{-7} cm³ per sec. (Mitra, 1947). Since the number density of O⁻ ions (during night time) is 2×10^{-5} times the electron density, the average concentration of Na⁺ ions is 6×10^4 per cm³. This value of the concentration of Na⁺ ions is obviously more acceptable than the one deduced above by the process (5). The total number of ions within the layer of thickness $4H$ is then 1.2×10^{12} ions per cm² column.

Thus we arrive at the conclusion that the number density of Na⁺ ions in the F-region must be at least of the order 6×10^4 per cm³, in order that the observed intensity of the D-lines in the night air-glow may be produced. This number constitutes about 10^{-5} of the total number of atmospheric particles in the region concerned.

Intensity variation throughout the night.—The intensity of the D-lines has been found to remain fairly constant throughout the dark hours of the night. Hence, if there be no means of replenishing the Na⁺ ions lost by neutralization, the initial number of ions has to be so high that the number lost during the dark hours is but a small fraction of the initial number. Now, the total number of transitions per cm² column throughout the night (say, 8 hrs.) is of the order 2.5×10^{12} . But, the number of Na⁺ ions per cm² column as deduced above is only 1.2×10^{12} . This number is obviously quite inadequate to maintain the required constancy of the radiation intensity. One is thus forced to assume that some process of re-ionization of the neutralized Na-atoms must be operative during the dark hours of the night. Such a process may be impact of particles of some sort incident on the atmosphere from outer space.

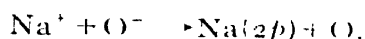
CONCLUDING REMARKS

In the light of the discussions given above, we may now draw the following conclusions regarding the distribution of sodium in the upper atmosphere, and also on the possible processes of emission of the D-lines.

It appears that sodium is concentrated in two layers: one in the 35 to 65 km. region, and the other in the F-region of the ionosphere. The former consists of neutral sodium atoms and the latter of ionized atoms (ionized by solar ultra-violet radiation). Alternatively, we may imagine that sodium in the upper atmosphere spreads from a height of 35 km to the F-region of the ionosphere. At lower altitudes sodium exists in the neutral state. The percentage of ionized sodium atoms increases with height and at altitudes above 100 km. all the sodium atoms are ionized.

The twilight flash of the D-lines is due to resonance excitation of neutral Na atoms by solar radiation $\lambda 5893$.

The D-lines in the night air-glow are emitted in the process of neutralization of Na^+ ions in the F-layer of the ionosphere. Of the two possible processes of neutralization—radiative recombination with electron and mutual neutralization with O^- ion—the latter appears to be the more likely process. Thus



The observed intensity of the nocturnal lines leads to the conclusion that the average density of Na^+ ions in the F-region is of the order 6×10^4 per cm^3 , constituting about 10^{-7} of the total number of atmospheric particles in the region. In the region of emission of the twilight flash the concentration is much less (by several orders). It is clear that the sodium distribution in the atmosphere does not follow the general exponential distribution law of the atmospheric particles.

The concentration of 6×10^4 Na^+ ions per cm^3 as given above is sufficient to produce the required rate of emission initially. But, it is totally inadequate to maintain the same (with only a small decrease in the rate) throughout the dark hours of the night. There must, therefore, be some process by which the neutral Na atoms, produced are constantly re-ionized during the night. Such night time re-ionization may be imagined to be produced by impact of extra-terrestrial particles. These particles may also contribute partly to the D-line emission by exciting the neutralized Na atoms to the $2p$ -states.

Alternatively, one may imagine that sodium atoms are entering the upper atmospheric regions from inter-stellar space and are being excited by collision with atmospheric particles. This latter hypothesis has the advantage that it can explain the high concentration of sodium (high in relation to that expected from the exponential law) in the 250 km. region.

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