Metallic Emissions from the Upper Atmosphere

Photometry of twilight emissions from alkali metals gives information on upper-atmospheric processes.

Donald M. Hunten

Spectral analysis of light from the upper atmosphere discloses lines and bands from the familiar constitutents, oxygen and nitrogen. This is hardly a surprise, but what is surprising is to find that the yellow D lines of sodium are also present. The greatest intensity of these lines is found in twilight and dayglow, but sodium emission is also present from the normal night sky (*airglow*) and, most strange of all, from the aurora. Careful observations have also revealed lines from lithium, potassium, and ionized calcium in the twilight. All these elements are truly minor constituents, present in concentrations of less than one atom in 10^{10} atmospheric particles; it is only the remarkable sensitivity of the optical method that makes them observable at all.

The emphasis in this article is on twilight measurements, which are usually the most interesting because from them it is possible to make a thorough quantitative analysis. This is based on the fact that the lines are excited by *resonance scattering*: absorption of a photon from the sun, followed by emission, usually in a new direction. Further information can be derived by measuring the decrease of intensity as the twilight shadow sweeps through the emitting layer. The situation is illustrated in Fig. 1, which shows the light from the sun passing near the earth's surface at the sunset line *S*, and then impinging on the upper atmosphere in the region *A* that is being observed. The light scattered at *A* is observed at *O*. When the angle of solar depression (β) is 5 degrees or more, the lower atmosphere along the line of observation is shadowed well enough so that the faint light from the upper atmosphere can be observed without great difficulty. Even better shadowing may be obtained by observing near the horizon, above the sun, instead of overhead; however, reliable results are more difficult to obtain because of the more complicated geometry. Measurements are usually made with fast spectrographs or photoelectric spectrometers; more recently, a number of specialized photometers, incorporating interference filters and some means of subtracting the background of white light scattered by the lower atmosphere, have been used. Another device, the "Zeeman photometer," is described in the last section of this article.

As I have mentioned, twilight offers the possibility of a thorough experimental and theoretical analysis. Much of this analysis has been carried out for its own interest, with little prospect of relating the results to the rest of the atmosphere. It has been necessary to consider the depletion of sunlight by the large number of sodium atoms encountered by the light in its two passages through the sodium layer, as shown in Fig. 1. Attempts have also been made to explain the existence of free sodium atoms in the highly oxidizing environment of the upper atmosphere. The sodium abundance shows a large seasonal variation, with a minimum in summer in both hemispheres and a maximum 5 to 10 times larger in winter, with no detectable change in vertical distribution. It appears that this can only be explained by changes in the rate of supply of sodium, perhaps due to large-scale circulations of which we know very little.

The much fainter emission from lithium has been observed only in twilight, and was not discovered until, as part of the auroral program of the International Geophysical Year, a large number of fast spectrographs were placed in service. It appears that the first observations were of natural lithium, but in 1958 a huge enhancement was caused by lithium from a thermonuclear explosion over Johnston Island in the Pacific. A similar enhancement occurred in 1962, and smaller events of this kind have been observed from time to time. Other sporadic lithium enhancements seem to occur naturally, in contrast to the smoother variation shown by sodium; the most natural explanation is an influx of meteoric particles.

In contrast to the accidental discovery of lithium, potassium was found only after years of searching by more and more sensitive methods. The abundance thus found can be combined with the results for the other alkalis to give further information on the sources. The simplest interpretation is that most of the sodium and potassium come from the oceans, and most of the lithium, from meteoric particles.

Lines of ionized calcium have been observed, but only sporadically; it appears that the intensity is usually just below the threshold of detectability. It is therefore difficult to draw conclusions, but there is some evidence that the appearance of the lines is correlated with meteor activity.

Because the scattering of sunlight by atoms of alkali metals is a very efficient process, it is possible to produce easily visible trails by ejecting metallic vapors from rockets. Observations of these trails give information on winds, turbulence, and diffusion in the atmosphere at heights between 80 and 200 kilometers, and also permit measurement of the temperature at these heights.

Observational Work on Sodium

The yellow emission, later found to be the D lines of sodium, was discovered in the night airglow by V. M. Slipher (*1*). Many attempts to identify the emitting atom failed because of the extremely low dispersion available, but the measured wavelengths continued to suggest sodium. A definite result was finally obtained in 1938 by means of Fabry-Perot interferometers, which were used almost simultaneously by Bernard and by Cabannes, Dufay, and Gauzit. At the same time, Bernard discovered the twilight enhancement; the interferometric method was immediately applied to this emission as well. Figure 2 shows the two D lines, recorded recently at Kitt Peak by A. L.

The author is a physicist at the Kitt Peak National Observatory, Tucson, Arizona.

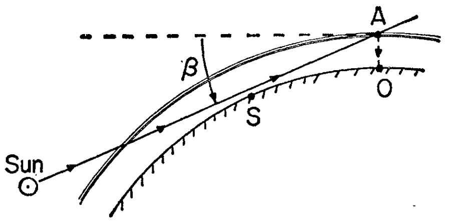

Fig. 1. Diagrammatic sketch of twilight, showing the sunlight coming in at a depression angle β, and being scattered at A to the observer O.

Broadfoot. Suggestions were made about the source of the sodium, and these were summarized by Chapman in an article which is still remarkably up-to-date after 25 years. Chapman was the first to suggest and discuss the chemical reactions responsible for maintaining free sodium atoms; with changes in details, the description is still in use.

The intensity of the sodium emission in twilight, if observed in the zenith, typically follows the course shown in Fig. 3. After sunset, a minimum intensity occurs when the angle of solar depression is about 2 degrees, because the incident sunlight must traverse a very long path through the sodium layer, losing much of its intensity at the wavelength required to excite the D lines before it reaches the point observed. This minimum can be observed only by means of special equipment able to discriminate against the very strong white light present at this time; with most spectrometers or photometers measurements cannot be made until the solar-depression angle is 4 or 5 degrees. At about 6 degrees the shadow of the lower atmosphere begins to encroach on the layer, and the intensity of the sodium emission starts to fall. The decrease is most rapid at slightly less than 8 degrees, and the descent to the nightairglow level is essentially complete at 10 degrees. At temperate latitudes, the steep part of this decrease occupies little more than 10 minutes; it is not surprising that early workers referred to a sodium "flash," bounded at one end by this rapid decrease and at the other by the obliteration of an observable emission by the intense continuum. The impression of an almost instantaneous disappearance also led to some difficulties with the "zenith-horizon" method of locating the sodium layer. This ingenious triangulation scheme consists of observing first in the zenith and then near the horizon above the sun (for an evening twilight). Comparison of the two curves permits deduction of a layer height and a "screening height." Unfortunately, it is very difficult to pick out matching points, and the difficulty is increased by the fact that the horizon intensity is about three times the zenith intensity. Thus, many results obtained with this method are in error, suggesting that the shadow is cast by a screening layer at a height of 40 kilometers or more. When correctly calculated, the screening height turns out to be 25 or 30 kilometers, as would be expected from ozone absorption and molecular scattering of the sunlight in its long twilight path.

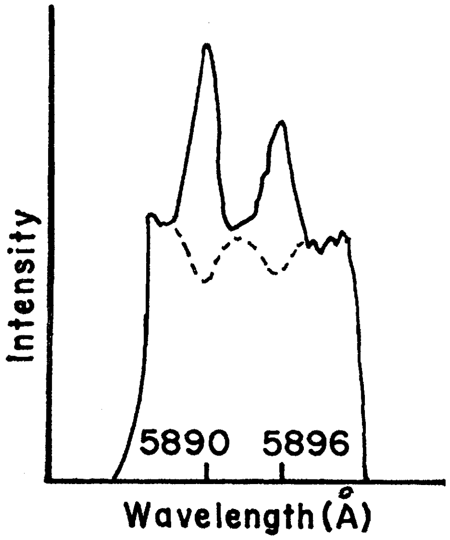

Fig. 2. A short twilight spectrum, showing the two D lines of sodium. The background solar spectrum is superposed in dotted lines. The tracings were made at Kitt Peak by means of a scanning photoelectric spectrometer.

The vertical distribution of the sodium can, in principle, be calculated from the curve of Fig. 3. In the simplest case, the shadow would be cast by a sharp screening layer; knowing the height of this layer, one could find the shadow height corresponding to each depression angle and replot the intensities as a function of the height. Differentiation of this curve would then give the vertical distribution. In practice, the shadow is fuzzy enough so that this derivative is only a first approximation. However, the magnitude of this effect can be calculated, and the broadening caused by it can be removed from the observed curve to give the required result (*2*). Some caution is necessary in using vertical distributions found in this way (Fig. 4), but the main features are certainly correct, and highly consistent results are found from night to night. The peak of the sodium layer is usually found close to 90 kilometers, although considerable fluctuation occurs on some occasions. Differences in the results are mostly due to the use of different methods of analysis; there is no real evidence for any systematic variation in height, either with time or with geographical position.

The height of the nightglow sodium layer has been measured by flying a photometer through it aboard a rocket. The nightglow layer bears a remarkably close resemblance to the layer found in twilight.

Chemical Reactions

The chemistry of sodium in the upper atmosphere has been discussed by a number of authors, following the original suggestions of Chapman (*3*). Presumably the other alkalis will behave in the same way, but differences will be caused by the differing reaction rates. It is assumed that sodium atoms are liberated from the compounds in which they arrive in the upper atmosphere either by solar radiation or by reaction with atomic oxygen, an important constituent at these heights. The subsequent reactions will usually be with the atomic and molecular oxygen, not with the original molecular partners. The most important reactions are thought to be

$$\mathrm{Na} + \mathrm{O_2} + M \rightarrow \mathrm{NaO_2} + M \qquad (1)$$

$$\mathrm{Na} + \mathrm{O_3} \rightarrow \mathrm{NaO} + \mathrm{O_2} \qquad (2)$$

$$\mathrm{NaO_2} + \mathrm{O} \rightarrow \mathrm{NaO} + \mathrm{O_2} \qquad (3)$$

$$\mathrm{NaO} + \mathrm{O} \rightarrow \mathrm{Na} + \mathrm{O_2}. \qquad (4)$$

The third body, M, in reaction 1 is required to permit the energy liberated to be carried away. The concentrations of O_2, O_3, and third bodies all fall off rapidly with increasing height; therefore, the rates of the oxidation reactions 1 and 2 do the same thing. In the region of interest, the concentration of atomic oxygen increases with height, and therefore so do the rates of the reduction reactions 2 and 3. Using estimated reaction rates for the four reactions, one can calculate that the fraction of the total sodium in the free form should be one-half at 90 kilometers, and that a peak should exist at the same height, in perfect agreement with observation. Above this height, nearly all the sodium is free; below, the rate of oxidation grows so rapidly that the amount of free sodium decreases again, even if the total amount is increasing. The resemblance to the observed distribution (Fig. 4) is so striking as to inspire considerable confidence in this picture. For the other alkalis, a

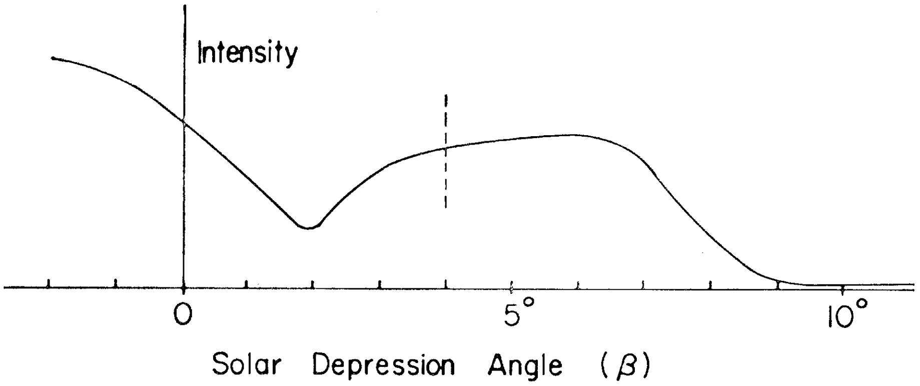

Fig. 3. Intensity of the sodium emission during and just before twilight. The dip near 2 degrees is caused by resonance absorption as the sunlight traverses the sodium layer horizontally; the decline around 8 degrees is due to the shadow of the earth and of the lower atmosphere. The part to the left of the dotted line is difficult to observe and was sketched in largely from theory.

layer of the same shape, but possibly lying at a different height, would be expected. According to the observations, the difference in the heights of the sodium and the potassium layers is small; the difference in the heights of the sodium and the lithium layers is about 11 kilometers.

Observation of the dayglow reveals a sodium concentration that may be as much as 10 times greater than the amount observed in twilight. Although there are some conflicting measurements, this effect appears to be real. Perhaps it can be explained in terms of diurnal changes in the ratio of O, O_2, and O_3; in addition, direct dissociation of sodium oxides by sunlight may play a part.

There is one other observation that does not fit into this picture. The lifetime of a vapor trail released from a rocket above 100 kilometers should be limited by spreading alone, since chemical reactions are slow and ionization is even slower. The rate of spreading can be observed by photographing the trail; the observed rate of decrease in number of atoms along the line of sight has been found to be more rapid. However, the interpretation of these measurements is very difficult and an error may have crept in (*4*).

It is likely that the sodium emission in the night airglow is closely related to the cycle of reactions 1–4. In fact, it is not impossible that reaction 4 is sufficiently exothermic to leave the sodium atom in the excited state. Definite laboratory evidence is lacking, but the available results suggest that the energy is not quite enough. However, in the atmosphere the NaO has just been formed by reaction 2 or reaction 3, and it may well be in an excited vibrational state, with enough energy to make up the difference. There is no real doubt that the supply of energy is from the recombination of atomic oxygen, whether by these reactions or by some other path (*5*).

Alkali atoms are readily ionized by solar ultraviolet radiation, and the required wavelengths can penetrate to the heights at which these atoms occur. The mean lifetimes before ionization are about half a day for sodium and potassium and 1 hour for lithium. It has never been possible to observe any of the predicted consequences of a high degree of ionization; thus, it appears that a rapid process of recombination must operate. Possibly this begins with a chemical reaction such as

$$Na^+ + O_2 + M \rightarrow NaO_2^+ + M. \quad (5)$$

The molecular ion can then react with an electron in a dissociative recombination, known to be a rapid process (*6*).

Abundances of the Alkali Atoms

The atomic constants are well enough known so that the intensity of light scattered from a known number of atoms can be calculated with considerable accuracy. The biggest uncertainty is in the effective intensity of sunlight, especially in the case of sodium and potassium. These elements are abundant in the sun, as well as in the earth, and absorb much of the light that would otherwise be available to excite the twilight emission. It is found that a sodium or a potassium atom scatters slightly less than one photon per second; a lithium atom scatters about 16 photons per second, the larger figure being due mainly to the essential lack of absorption of the exciting light at the sun. Total abundances can then be found from the observed "plateau" intensities (Fig. 3); they vary between 10^9 and 10^{10} atoms per square centimeter for sodium, and 10^6 and 10^7 (occasionally higher) for lithium. The few measurements of potassium that have been obtained give about 10^8 atoms per square centimeter. More interesting, and more difficult to measure, are the concentrations, such as are shown in Fig. 4 (*6*). The peak values for sodium and potassium, respectively, are 1200 and 25 atoms per cubic centimeter, to be compared with 5×10^{13} for the whole atmosphere at 90 kilometers. The figure for lithium is only 2 atoms per cubic centimeter in an atmosphere of 5×10^{14}. We are truly dealing with minor constituents here; it is only their remarkably high light-scattering ability that makes them observable at all.

The amounts of sodium found are sufficient to cause a considerable depletion of the incident sunlight under twilight conditions; this effect is usually called "resonance absorption" and is closely related to the "self-absorption" sometimes found in laboratory light sources. The most striking effect, although very difficult to observe, is the notch at about 2 degrees of solar depression in Fig. 3. However, there can also be a considerable reduction of the plateau intensity and a distortion of the shadow cutoff used for finding vertical distributions. All these effects have been the subject of detailed calculations, which have taken into account the line shapes, as affected by thermal motions and hyperfine structure, and multiple scattering of photons as they wander through the sodium layer. It is necessary to remember that the incident sunlight must pass through the sodium layer on the day side before it grazes the earth's surface on its way to the twilight layer (Fig. 1). Another interesting result is that the intensity ratio of the doublet (the D lines of sodium) is considerably depressed below its normal value of 2, partly because the two lines are unequally absorbed in the sun, but also because the strong line is more strongly affected by resonance absorption. Again, this ratio is difficult to observe, but the results are in satisfactory agreement with calculation (*7*).

Because of the extreme rarity of sodium atoms, the D lines would not be expected to appear in the spectra of auroras; particle bombardment should give relative intensities that are nearly the same as the relative concentrations. The presence, suspected for some time, of the lines in the spectra of auroras of a certain type has recently been confirmed (*8*). These are auroras of a distinctive form, with red lower borders, known to occur at unusually low heights of 80 to 90 kilometers. The most likely excitation mechanism is a transfer of energy from a molecule of N_2 or O_2 in an excited vibrational state.

Variations and Origins

The sodium abundance shows a remarkably large seasonal variation, illustrated in Fig. 5 for stations at 52° north and 44° south geographic latitudes (at Saskatoon, Canada, and Christchurch, New Zealand). The intensities obtained at Christchurch are somewhat higher than those obtained at Saskatoon; this difference is probably not real, since it is very difficult to transport an intensity calibration over the long distance between these two stations. The principal feature to be noted is that the variations occur according to season, not date. Such a seasonal variation cannot readily be explained as due to changes in influx of meteoric material, unless highly artificial assumptions are made (*9*).

The intensity variation of the sodium emission in nightglow appears to be very similar to the abundance variation deduced from twilight measurements, although it is much more difficult to measure. This similarity would be expected if the excitation is a by-product of the chemical cycle, as suggested earlier.

The seasonal variation in abundance deduced from twilight measurements could be explained if the height of maximum concentration varied by some 15 kilometers during the year; the chemical equilibrium could conceivably change by this much. However, height measurements disclose no variation greater than about 3 kilometers, and therefore this mechanism also must be discarded.

The seasonal variation bears some resemblance to that found for ozone at high latitudes, which was unexplained for many years. It is now believed that the ozone is transported to relatively low levels during the violent meteorological events of the polar night. It seems reasonable to assume that small salt particles could be transported upward by a similar mechanism. Proof of this mechanism is even more difficult to find for sodium than it was for ozone, but there is some prospect that a test of the hypothesis will soon be possible.

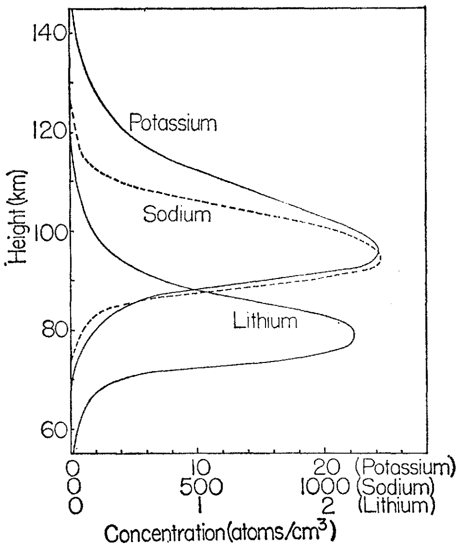

Fig. 4. Vertical distributions of the three alkali metals measured at Saskatoon, Canada. Note the three different concentration scales.

Further information on the origins can be gained by studying the relative abundances. According to the chemical picture presented earlier, the atoms are essentially all in the free form above the peak of the layer; thus, two layers at the same height, like the sodium and potassium layers, can be compared directly. Lithium remains dissociated to much lower heights, and a correction must be made: the observed abundance must be divided by 6. It is possible that the observed ratios should also be corrected for fractionation between the source and the atmosphere; however, no mechanism has been suggested, and it is simpler to assume that the correction is negligible. The observed ratio of sodium to potassium is close to the value in sea water; in the other likely sources the potassium abundance is about 5 times greater than the sodium abundance. This gives strong support to the marine origin suggested above (*6*).

Systematic observations of lithium require an unusually sensitive instrument; an instrument of this kind has so far been available at only one station. For lithium, the abundance variation is of an entirely different character, giving the impression of a constant background value with sharp peaks superimposed, each lasting 3 or 4 weeks. Contaminations with large amounts of lithium from thermonuclear explosions, though interesting in themselves, have hampered these observations. However, the largest maximum appears to recur in November, and a smaller, shorter one has been observed 2 years out of 3 in early February. A possible correla-

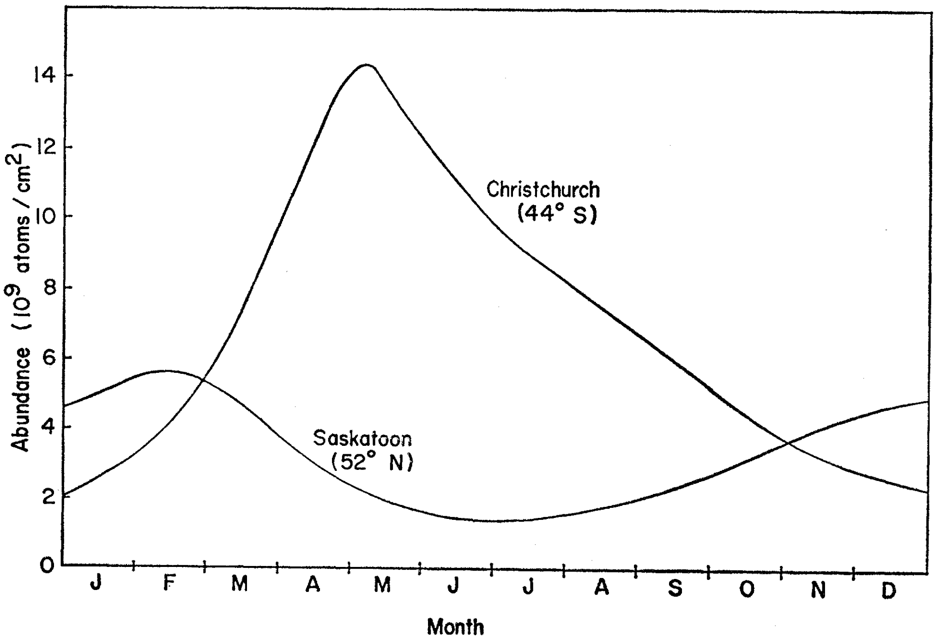

Fig. 5. Sodium abundances in the Northern and Southern hemispheres, showing the approximate 6-month shift (*9*). As mentioned in the text, the difference in the abundance scales is probably not significant.

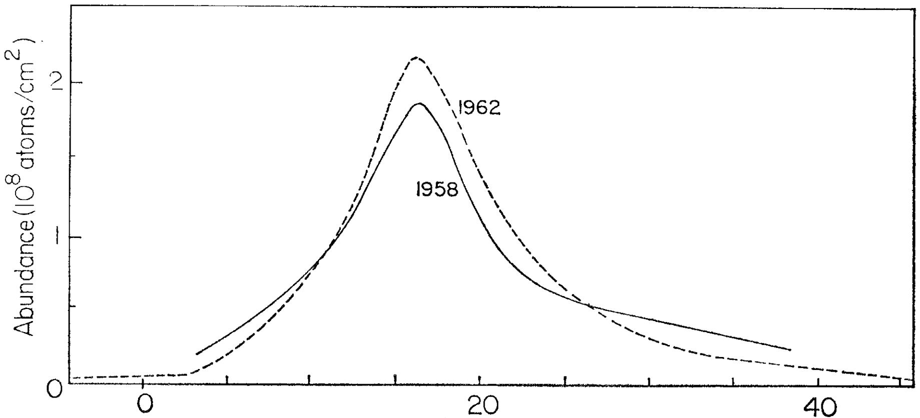

Fig. 6. Variation of lithium abundance during the enrichment events of 1958 and 1962, believed to have been produced by thermonuclear explosions at a height of 50 kilometers.

tion has been suggested with "dust showers" observed by micrometeorite detectors on satellites.

Between these peaks, the lithium abundance is still much too large to be consistent with a marine origin, even after the correction described has been made. However, the lithium content of meteoritic material is large enough to account for the observed lithium, while meteoritic material would contribute no more than a small percentage of the total abundance of sodium (*6*).

The two largest events of artificial enrichment observed to date are shown in Fig. 6. The similarity is remarkable, especially since the two were observed from different hemispheres and different latitudes. Both appear to have been produced by rocket-borne thermonuclear bombs exploded at a height of about 50 kilometers above Johnston Island. A much smaller effect was observed after the Soviet 60-megaton explosion of 1961 in the lower atmosphere. Still smaller effects were produced by the two large explosions above the atmosphere, one set off by the United States and one by the U.S.S.R. These five events are the only ones known to have produced an observable effect on the atmospheric lithium (*6, 10*).

Rocket Trails and Dayglow

Less than a kilogram of alkali metal vapor is enough to produce a long trail easily visible in twilight. The trail can be photographed from two or more stations, successive exposures being made to permit deduction of the motion and expansion of the trail. Valuable information on winds and turbulence at different heights has been obtained in this way. Temperature is found by measuring the transmission of the scattered light through a cell containing the same vapor, and then calculating the line width. When such measurements are made on a sodium trail, there is serious interference from the natural sodium, just as there is in the natural twilight. It is even possible to find the shadow of the natural layer on the trail. Interesting though this may be, it detracts from the accuracy of the other measurements, and lithium or potassium are preferred for this reason (*11*).

The only observations of sodium dayglow, so far, have been made from the ground by a "Zeeman photometer." The sky light is scattered by a sodium-vapor cell, and the scattered light is measured by a photomultiplier. Even when the best available interference filter is used in the system, the intensity of background light is still overwhelming. However, when a magnetic field of a few thousand gauss is applied, the sodium light is no longer scattered, while the parasitic (or background) intensity is only slightly affected. When allowance has been made for this small effect, the difference between the two signals gives the intensity of the sodium dayglow. One would expect this to be somewhat higher than the twilight intensity, because of the different conditions of illumination. However, the observations show an intensity as much as 10 times higher. On the other hand, other methods of finding the sodium abundance in daytime by the absorption effects give results that are in agreement with the twilight measurements. The discrepancy is disturbing, and a more direct determination, perhaps from a rocket, would be desirable (*12*).

Future Prospects

We have seen that, although considerable effort has been devoted to the observation of atmospheric sodium and the interpretation of the results, there is still some doubt about the basic facts. Twilight measurements are consistent among themselves, but they are contradicted by some measurements made in the daytime. The solution is likely to be found through the use of rocket-borne photometers observing from above the bright lower atmosphere. As a bonus, the photometer will be able to measure the height of the sodium as it penetrates the layer. Thus, if additional sodium is present in the daytime, its location in the atmosphere may provide a clue to the explanation of its presence.

The seasonal variations in abundance are large at temperate latitudes, but they would be expected to be much smaller near the equator. A study of the form of the crossover would be interesting. If the sodium really is injected into the upper atmosphere during the polar night, correlation with meteorological observations may be possible, and the migration of the sodium toward the equator might be observable.

Systematic observations of potassium will give a better estimate of the potassium-sodium abundance ratio and will show whether, as expected, the seasonal variations for the two elements are similar. If this expectation is confirmed, the hypothesis of a marine origin will be strengthened; if not, it may have to be abandoned. Similarly, further observation of lithium may show whether meteoric influx really is important; these observations should be much easier now that the chances of artificial contamination are greatly reduced.

If all the expectations are confirmed, perhaps the study of these twilight emissions, which is now carried out mainly for its intrinsic interest, will finally begin to contribute information about the upper atmosphere itself.

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News and Comment

Environmental Health: Taft Center in Cincinnati Has Been the PHS Mainstay in Pollution Research

Cincinnati. In mid-June the Health Physics Society met in this city, which, because of the presence of the Robert A. Taft Sanitary Engineering Center, can be called the spiritual home of health physics. Members of the Health Physics Society are scientists, engineers, and physicians concerned with the effects of radiation on humans, one of the newer problems of pollution of the environment. And such pollution has been the main subject of interest at the research facility established here by the Public Health Service just over a half century ago.

Until the end of World War II, the center in Cincinnati was a small operation devoted almost exclusively to water pollution problems. Then after the war, for a number of reasons—sheer growth of population and industry, emergence of atomic energy with its peculiar problems, increasing realization that smog is not only a nuisance but a menace in many cities, the so-called chemical revolution with its by-product problems relating, for example, to pesticides and detergents—there occurred what might fairly be called a pollution explosion. Taft Center researchers moved into these new areas, particularly as Congress, in a rather random way, recognized the new dimensions in public health problems with new laws and new funds.

The PHS center was established in 1913 specifically to study the Ohio River which flows by the city's doorstep. By the turn of the century the beautiful blue Ohio and many of its tributaries were badly polluted with sewage and wastes from factories and mines in the Ohio basin. The PHS choice of Cincinnati, which is roughly halfway down the river, seems to have been influenced by the presence in the city of an old marine hospital operated by PHS. The center's name, orginally, was the descriptive Stream Pollution Investigations Station.

The center's current director, Harry P. Kramer, notes that primary emphasis at the station was given to establishing the principles of natural purification of water in streams, about which not a great deal was known then, and to developing treatment systems for polluted water. From the outset, says Kramer, the station was successful in attracting highly competent men from different fields—biologists and microbiologists, chemists, engineers, and physicians—thereby setting the pattern for the approach to pollution problems that has been followed at the center ever since.

The researchers soon moved on from their exclusive study of the Ohio to a concern with the problem of stream pollution in general. Work done at the Cincinnati center soon found its way into textbooks, and the station has exercised a heavy influence in sanitary-engineering education in American universities.

During World War II, the station, like most other government agencies, was called on to deal with immediate problems, such as developing waste treatment systems for military installations. One of the first intimations of the coming diversification of pollution problems, came when the station was consulted on the disposal of radioactive wastes. Then in the late 1940's, research began, in a small way, on various kinds of pollution of food and milk. In 1949 the name of the station was changed to the Environmental Health Center of the Public Health Service, and plans were developed for construction of a building to fit the center's new duties and personality.

The building was occupied in 1954, and the center was renamed the Robert A. Taft Sanitary Engineering Center, after Ohio's senior senator who had died a short time earlier. The reversion to "sanitary engineering" in the name seems to have been decided upon as a means of emphasizing the tie between the center and sanitary-engineering education in the universities.

The trend toward bigness at the center was reflected organizationally by a setting up of divisions corresponding to the four main categories of work at the center: Division of Water Supply and Pollution Control, Division of Air Pollution, Division of Radiological Health, and Division of Environmental Engineering and Food Protection.

The center and all its divisions are subordinate to the PHS Bureau of State Services and are under the super-