Solar variability and the lower atmosphere

Abstract
Mechanisms possibly connecting solar activity to meteorology of the lower atmosphere are reviewed. Besides direct variations of solar visible emission, solar-related fluctuations in some aspect of cloudiness could be important. Any such variations in cloudiness are likely to be related to variations in production of ionization near the tropopause by galactic cosmic rays, the only geophysical phenomena unconnected with upper atmospheric processes known to have a striking (negative) correlation with solar activity. Such a connection might involve a dependence of sulfate aerosol formation on ionization and in turn a dependence of cloud radiative properties on variations of the aerosol particles' action as cloud condensation nuclei.

1. Introduction
For some time now I have been attempting to revise the AMS statement on solar effects on weather. However, little progress was made until I received an invitation to present my views on the connection between solar activity and the lower atmosphere at a session on "Solar Variability and Meteorological Response" at the 55th AMS Annual Meeting in Denver, January 1975. To prepare that paper it was necessary to develop some ideas as to where physical connections between solar variability and meteorology are most likely to be found. I hope to share these ideas further through this written version of my oral paper.

The extensive literature noting apparent solar-weather (or climate) relationships (as recently reviewed, e.g., by Wilcox, 1975) suggests that there probably are some connections between changes on the sun and changes in the lower atmosphere. For example, the recent results of Wilcox et al. (1974) relating zonally-averaged vorticity to solar sectors appear to be statistically significant (Hines and Haley, 1975). On the other hand, there are strong theoretical arguments for rejecting most suggestions as to how solar activity could affect the lower atmosphere. It is not my intent here to judge the validity of apparent observational relationships between solar activity and the lower atmosphere, but rather to indicate which physical processes in the lower atmosphere would most likely be involved in the explanation of such relationships.

The gross features and time variability of the earth's atmosphere and near space environment have now been elucidated at all levels. In particular we know that particle precipitation and electric fields and currents, which are correlated with solar activity, strongly modulate the temperature and other processes of the atmosphere above 100 km. Many investigators have sought to tie this solar connected thermospheric variability with changes in the lower atmosphere. Two strong arguments can be made for the futility of such efforts. First, the energy variations in the regions established as responding strongly to solar variability are less than 10^4 the amount of energy required to influence the lower atmosphere. Second, most suggested coupling mechanisms would produce much greater effects on the stratosphere and mesosphere than on the lower atmosphere. However, no such large variations of stratospheric or mesospheric structure in response to solar variability have been identified.

As already noted, it is the intent of this paper to discuss those physical processes in the lower atmosphere whose further study might provide a key for interpretation of connections between solar activity and weather and climate. A guiding principle I have used for identifying these processes can be stated as follows. Such mechanisms should either involve processes in the lower atmosphere that have a large known variation with solar activity or they should involve energy relationships in the lower atmosphere that could have variations too small to have yet been detected and yet sufficiently large to possibly have observable effects. Thus, first we must look at the energy cycles of the lower atmosphere to identify the processes that by their variation with solar activity, if any, could significantly affect weather or climate. Second, we should ask if there are any phenomena in the lower atmosphere that already are known to vary with the solar cycle and see if they can be tied to any of the above climate-affecting processes.

In the next section I consider the energy processes of the lower atmosphere, asking in particular which linkages might be important for connecting solar activity to the lower atmosphere. It is concluded that only significant variations in the absorption of solar radiation or of the emission of infrared radiation by the lower atmosphere and earth's surface are capable of producing notable changes in the lower atmosphere. One possible cause of such changes might be changes in solar emission at visible or near infrared wavelengths, with amplitudes too small to have yet been detected observationally. A second possible cause might be changes in the distribution of cloudiness. This latter possibility would require some indirect linkage to solar activity. In the following section, I shall review the only lower atmo-

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er geophysical process that is known to have a large variation with solar activity. This is the ionization due to galactic cosmic rays. Then I speculate as to how this ionization might modulate aerosols that are effective cloud condensation nuclei and so modulate cloudiness in the upper troposphere. Evidently, further understanding of the role of ions in aerosol formation and the role of sulfate aerosol in determining upper tropospheric clouds is required to be able to verify or (as seems more likely) to disprove these ideas.

2. Energetics of the lower atmosphere

Mechanisms by which solar activity might affect the lower atmosphere fall into two groups: a) those involving some change of the heat budget and b) those involving a modulation of dynamic processes. Within each of these groups there are several distinct classes of mechanisms. For example, the heat budget could be modulated by changes in: 1) direct heat inputs; 2) tropospheric or surface reflective properties; 3) distribution of radiatively active constituents, e.g., O₃, NOₓ, CO₂, H₂O and hence absorption properties; 4) the hydrological cycle. Dynamic processes could be modulated by changes in: 1) momentum inputs; 2) reflection of waves leaving the lower atmosphere, e.g., by modifying static stability or mean thermal gradients in the stratosphere or above; 3) tropospheric baroclinicity, i.e., by modification of the meridional temperature gradient.

In considering these possible dynamic modulations, we note that all but the first require a prior change in atmospheric temperature, that is, a change in the heat budget, and so cannot be effected independently of heat budget changes. An input of momentum from the sun would have to be transmitted to the earth by solar particles which would be absorbed above 100 km. Again there are severe difficulties as to adequate magnitude and the need for momentum to be transmitted as a hydrodynamic wave through the stratosphere and mesosphere which make this suggestion unlikely. Dynamic and thermal processes in the atmosphere generally change together, because of the thermal wind balance between the wind and temperature fields. It consequently is not simple to distinguish, on the basis of an observed response, between dynamic and thermal inputs. In contrast to any possible dynamic modulations known to us, we shall see that some of the suggestions whereby solar activity might affect the lower atmosphere heat budget are not entirely implausible. Let us first review the gross aspects of the average heat budget of the earth-atmosphere system.

The basic drive is solar radiation, which for considering the lower atmosphere is adequately represented by 6000 K black body emission. Only 70% of the solar radiation is absorbed, the rest being reflected back to space, mostly by clouds, but also by the earth’s surface, atmospheric aerosols and molecules. The amount of back-reflection depends also on the concentrations of atmospheric gases absorbing solar radiation. The earth-atmosphere system returns this energy to space by infrared radiation with energy output equivalent to that of a 252 K black body. The infrared radiation comes from the earth’s surface and from the various atmospheric gases with infrared emission spectra, most notably from H₂O, CO₂ and O₃. The manner in which the incident solar and outgoing infrared are intercepted by gases is well illustrated by the figure on p. 7 of Goody’s (1964) text, here reproduced as Fig. 1. The effective earth-atmosphere radiating temperature is determined by the net amount of absorbed solar radiation. The temperature at the earth’s surface in turn depends on the altitudes at which the outgoing emission originates. Since the temperature of the effective emission level is fixed by the balance with the absorbed solar radiation, increasing the fraction of emission from higher (i.e., colder) layers increases the temperature at the earth’s surface. Both clouds and absorbing gases contribute to determining the height of the level of emission.

The fate of incoming solar and outgoing infrared radiation is summarized by consideration of the annual average gross radiation budget. In this budget are analyzed 1) the fraction of the incident solar beam that is absorbed by surface or atmosphere, or that is reflected by clouds or by earth’s surface, aerosol, and dust, and 2) the amount of infrared radiation that is emitted from the earth’s surface and that is emitted upward and downward by atmospheric gases and by clouds. A recent estimate of this budget from London and Sasamori (1971) is shown in Fig. 2. We particularly note the 0.3 of the solar beam that is reflected back to space, with clouds responsible for about 0.8 of this reflection. Clouds are also important in determining the net infrared emission or as discussed above the effective level of emission from the atmosphere.
The effect of varying global cloud cover and radiating height is illustrated by Fig. 3 from Schneider (1972). The model that was used to calculate surface temperature in this figure is one-dimensional, with a prescribed tropospheric lapse rate of $-6.5 \text{ K km}^{-1}$, a fixed stratospheric temperature of 218 K, and relative humidity profile held constant. Such models predict approximately a 1 K change in global mean temperature per 1% change in the solar constant. There is a large uncertainty in the magnitude of this result as a consequence of neglect of various important feedback processes whose contribution to surface temperature change is difficult to estimate. However, the relative sensitivity of surface temperature to changes in cloudiness vs changes in solar constant are reasonably estimated by the model.

Thus the figure shows that about an 8% change in cloud cover is equivalent to a 2% change in solar constant. The equivalence between cloudiness increase and solar constant decrease also could have been estimated by assuming a global cloud cover which reflects back 50% of the solar radiation incident upon it. Such an estimate, however, would have indicated that an 8% change in cloudiness is equivalent to a 4% change in solar constant. The decrease of infrared cooling with an increase of mean cloud cover compensates on a global mean basis for approximately half of the decrease of solar radiation absorption. The precise amount of compensation between changes in solar heating and infrared cooling for changes in cloud cover will depend on the altitude and latitude of the changing cloud cover. At high latitudes, and especially in the winter hemisphere, the compensating change in infrared cooling is greater than the change in solar heating. Figure 3 also shows that surface temperature is extremely sensitive to changes in cloud heights. A mere 0.5 km increase in cloud height increases surface temperature by as much as would an increase of about 2% in the solar constant. With increased cloud heights the mean effective radiating height is raised. Since the temperature lapse rate from this height to the surface is (assumed) unchanged, but the effective radiating temperature is fixed by the absorption of solar radiation, surface temperature must increase.

Thin clouds or aerosol layers can also have a significant effect on the earth-atmosphere heat balance. For example, the transmittance of direct solar radiation at Mauna Loa Observatory showed an order of 2% drop from years preceding to years following the Mt. Agung volcanic eruption (Ellis and Pueschel, 1971), presumably due to an increase of stratospheric aerosol (the eruption has been shown to have increased the loading of stratospheric aerosol by an order of magnitude). The scattering of a solar beam by stratospheric aerosol is 80% to 90% in the forward direction, so the reduction of effective solar constant associated with the Agung eruption would have been estimated by the model.

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eruption, if due entirely to increase of stratospheric aerosol, is estimated to be one to several tenths of a percent of the solar constant. If, however, the increase of stratospheric aerosol results in a modulation of cirrus cover at the tropopause, a possibility we shall raise later, it becomes difficult to make any estimate of the reduction in solar energy reaching the tropopause due to the eruption.

Stratospheric particles are characterized by sizes of order of 0.3 μm, or two orders of magnitude smaller than typical water or ice cloud droplets. Thus, a thin cloud of water droplets or ice crystals has a considerably different effect on the net radiation balance than does a layer of “typical” stratospheric aerosol. The results of calculations (Coakley \(^3\)) of solar heating and infrared cooling by a cirrus deck with a 20% infrared emissivity located near the tropopause in the winter hemisphere are shown in Table 1. The rates apply to the mean heating or cooling in the atmospheric column below the cloud. Net solar heating changes but little, but approximately 0.1 K/day less infrared cooling in the column below results with the assumed cirrus deck in place. It must be remembered, however, that the amount of heat required to produce a temperature change is generally more than that which would be required if all the heat input changed temperature alone. Some of the heat input is invariably balanced by redistribution by motions and by increased infrared cooling. The cirrus cloud also decreases significantly the surface infrared cooling (equivalent to about 5% of the global mean incident solar radiation), and this cooling would have to be included if long-term climate variation were being considered. To estimate the possible effect on winds, note that a mean 0.1 K decrease in the temperature difference between atmospheric columns in middle latitudes separated by 15° latitude would decrease the zonal mean westerly wind at the tropopause and between these two latitudes by order of 2 m/s. The example we have just discussed is given as an estimate of the minimum effect that might be detectable on a short-term basis. See Murray et al. (1974) for further references as to the radiative effects of cirrus clouds.

Surface temperature can also be changed by changes in the lower stratosphere. Significant radiative coupling of the lower atmosphere to the stratosphere occurs through the emission and absorption of infrared radiation by ozone, carbon dioxide, and water vapor, and by the absorption of visible radiation by ozone in the Chappius bands near 0.6 μm or by NO₂ in the near ultraviolet. A decrease of temperature or ozone concentration in the lower stratosphere will decrease the infrared radiation transferred downward from the stratosphere to the earth’s surface and so lead to surface cooling. On the other hand, decrease of the total ozone column content also leads to increased visible radiation reaching the surface and for this reason, net heating. Manabe and Wetherald (1967) calculated global mean radiative equilibrium temperature profiles for polar, midlatitude, and equatorial profiles. The polar profile with the most ozone in the lower stratosphere gave a 1 K warmer surface temperature than the reference midlatitude profile. Similarly, the equatorial profile with the least amount of ozone in the lower stratosphere gave a 1 K colder surface temperature than the reference case. Apparently in their calculation, the surface temperature responded more to the changes in infrared than to the compensating changes in visible flux. Other more recent calculations (Coakley \(^3\)) indicate that the net surface change is as sensitive to changes in the vertical distribution of the ozone as to the change of total ozone.

To summarize, there are several processes determining the heat budget of the earth-atmosphere system which could have a correlation with solar activity that has not yet been detected. As the simplest possibility, solar radiation emitted at wavelengths which can reach the lower atmosphere might be variable, provided the magnitude of these variations is less than the order of 1% indicated as an observational upper limit. If this were the case, an explanation for atmospheric changes would have to be sought more in solar than in atmospheric physics.

There are also, however, several atmospheric components contributing to the net heat budget below the tropopause that also could have as yet undetected variations with solar activity. Furthermore, the observational constraints on these variations are sufficiently loose that changes in the heat budget at the surface larger than would be expected from the upper limit on the variation of incoming solar radiation is not impossible.

Distribution of global cloudiness and especially cloud top height certainly should be considered. A 5–10% change in thick cloud cover or a 0.5 km change in cloud heights with solar activity would probably have gone undiscovered up to now. Changes of thin (cirrus) cloud cover in otherwise cloudfree areas must also be regarded as a prime possibility for changing surface heat budget. Finally, changes of the vertical distribution of stratospheric ozone, temperature, and aerosol loading with solar activity could also simulate (or obscure) effects of solar constant variation.

### Table 1. Mean IR cooling rates (K/day) for cirrus cloud between 10 and 12 km and with 20% emissivity.

<table>
<thead>
<tr>
<th>Lat. (Jan.)</th>
<th>Clear</th>
<th>Cloud</th>
<th>Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>30°N</td>
<td>1.66</td>
<td>1.49</td>
<td>0.17</td>
</tr>
<tr>
<td>45°N</td>
<td>1.24</td>
<td>1.12</td>
<td>0.12</td>
</tr>
<tr>
<td>60°N</td>
<td>0.999</td>
<td>0.907</td>
<td>0.09</td>
</tr>
</tbody>
</table>

Changes in solar heating ≈ 0.01 K/day.

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3 Personal communication.

### 3. Cosmic ray ionization

In the previous section we have identified several processes that might vary with solar activity. Unless changes in the incident visible or near infrared radia-
ionization are involved such a variation would not be linked directly to solar activity. Rather, it would depend on the existence of some intermediate process which does have an obvious connection to solar activity. The lower atmosphere process most strikingly dependent on solar activity is ionization by galactic cosmic rays. We shall now review what is known about this ionization and its dependence on solar activity as a prelude to speculating as to how this ionization might influence the lower atmosphere heat budget.

Most of the ions between 1 km and 50 km are generated by the high energy particles known as cosmic rays. Decay of radioactive elements in the surface rocks also produces ionization near the ground, whereas above 50 km, solar electromagnetic radiation, e.g., x-rays, Lyman-α, etc., becomes the dominant source of ionization. Cosmic rays are mostly protons and consist of two components, galactic and solar cosmic rays. The galactic cosmic rays, which we shall discuss first, arrive constantly with a mean flux at the top of the atmosphere of several tenths cm$^{-2}$ s$^{-1}$ and a mean energy per particle of order 0.1 MeV (i.e., ~1 GeV). The net effect of the stopping of these particles by the atmosphere is the production of order of $10^7$ ions per particle and comparable molecular dissociation, albeit there are first produced a multitude of the ephemeral particles best known to high energy physicists. The greatest rates of ionization occur at the pressure level where an average particle has expended all but $1/e$ of its energy. This generally lies near the tropopause in high latitudes. Particles of energy less than several GeV cannot reach equatorial latitudes. Consequently, ionization rates near the equator maximize in the middle troposphere and are lower than rates in high latitudes. The inverse dependence of these ionization rates on solar activity, which we shall further discuss, is nicely illustrated by the variation of the differential proton spectrum with solar activity shown in Fig. 4, from Neher (1967). Greatest fluxes occur at several hundred MeV during low solar activity (e.g., in 1954 and 1965) whereas there is little flux at less than 1 GeV during high solar activity (e.g., in 1958). This flux variation would be expected to modulate most greatly ionization rates of the high latitude stratosphere.

Figure 5 from Neher (1971) illustrates this modulation over a solar cycle at Thule, Greenland. The solar cycle variation is a factor of two at 50 mb and order of 30% near 200 mb. Note that this figure gives ionization rates per unit mass. Maximum rates per unit volume occur between 150 and 200 mb. These rates of galactic cosmic ray ionization have been measured using balloons (cf. also, Anderson, 1973). The number of flights have apparently been inadequate for resolving variations of
cosmic ray fluxes on time scales shorter than the solar cycle. On the other hand, surface monitoring of cosmic ray fluxes, e.g., by neutron counting, has been carried out for many years with fine time resolution, so that not only solar cycle variation but also shorter term fluctuations down to time scales of hours or less have been evaluated. Analysis of C¹⁴ content in tree rings allows extension of the cosmic ray history back several thousand years (e.g., Suess, 1968).

The most notable disturbances on short-time scales are known as Forbush decreases, which last for several days to several weeks and are highly correlated with the occurrence of geomagnetic storms (e.g., Lockwood, 1971). Other correlations that have been noted include a 27-day periodicity and an asymmetry between hemispheres depending on solar sector magnetic field (e.g., Wilcox, 1968; Yoshida et al., 1971). Unfortunately, surface measurements detect only the high energy tail of the incident cosmic ray fluxes which apparently are not nearly so sensitive to solar activity as are the lower energy particles that dominate the maximum rates of ion production near the tropopause.

In contrast to the galactic cosmic rays, solar proton events occur only sporadically during “events” of a few days lifetime. Their frequency but not intensity correlates positively with periods of sun-spot maximum. The mean energy of solar proton events per particle is usually at least an order of magnitude less than that of galactic cosmic rays (Zmuda and Potemra, 1972). There is, to be sure, a much greater number of incident particles during solar proton events than due only to galactic cosmic rays. Consequently, maximum molecular dissociation and ionization rates are produced in high latitudes near the stratopause or above, where they disrupt radio communication. During the largest events, which occur every few years, as much ionization may occur as produced by an entire year of galactic cosmic rays. Even less frequently there are some particles with enough energy to reach the surface. These are known as solar cosmic ray events.

However, the solar cosmic ray ionization below 20 km, even during the most energetic events, may not significantly exceed the mean galactic cosmic ray ionization rates (Zmuda and Potemra, 1972) and so in the mean is apparently a much less important source of ions for the troposphere and lower stratosphere than is ionization by galactic cosmic rays.

The relationships between solar and galactic cosmic rays and solar activity are understandable in general terms but some details are still not well understood. Solar cosmic rays are associated with, and apparently generated by, solar flares. Galactic cosmic rays, on the other hand, are assumed to have a homogeneous spatial distribution of fluxes outside the solar system. Current models of the propagation of galactic cosmic rays with energies of several hundred MeV or greater (e.g., Jokipii, 1971) consist of a balance between advection outward from the sun by solar wind, and transport toward the sun by random diffusion processes depending on scattering of particles from magnetic turbulence in the solar wind. For steady conditions, the rate of spatial decay towards the sun consequently should vary as the solar wind speed divided by the appropriate diffusion coefficient. Increases in solar wind or decreases in the diffusion coefficient, according to these models, implies increases in galactic cosmic ray fluxes in the vicinity of earth. It has been suggested that Alfvén waves in the solar wind may be a major mechanism for scattering galactic cosmic rays; cf., e.g., Lee and Leerhe (1974) who give a derivation of the cosmic ray diffusion coefficient in terms of the Alfvén wave power spectrum. This model probably accounts for the mean decrease of cosmic ray fluxes approaching the sun. There is, however, little observational evidence that solar wind velocities or solar wind turbulence vary significantly with the solar cycle (Pomerantz and Duggal, 1974). Thus it is not clear whether the diffusion-advection balance can explain variations of cosmic ray fluxes with the solar cycle, or with solar activity in general. On the other hand, Barouch and Burlaga (1975) have presented convincing observational evidence that Forbush decreases occur with the arrival of the cosmic ray particles in the solar wind, which originate either in flares or “M-regions.” The latter are now identified with coronal holes (e.g., Hundhausen, 1972; Krieger et al., 1973) where solar magnetic field lines are open to space. Barouch and Burlaga considered but two years of data, so that solar cycle dependence of their blobs is not evident. If, however, these blobs are an important trigger mechanism for geomagnetic storms, their frequency presumably must vary with the solar cycle. Such a variation would explain the minimum galactic cosmic ray intensities at high solar activity.

4. Formation of stratospheric aerosols and cloud condensation nuclei in the upper troposphere

Up to now I have more or less restricted myself to hard science. To recapitulate two major points: a) climate is sensitive to changes in the absorption or reflection of visible or infrared radiation by atmospheric gases or particles, b) there is a pronounced solar cycle modulation of the dominant source of tropospheric and stratospheric ionization. If there are no significant variations of visible radiation emanating from the sun, then it would seem that any other possible relationship between solar activity and climate must involve some connection between the above points.

Several authors in the past have put forth suggestions as to possible linkages between cosmic rays and atmospheric structure. Ney (1959) pointed out the variation of galactic cosmic ray ionization with solar activity and suggested this variation should be considered in looking for solar-weather connections, especially with regard to electrical phenomena such as thunderstorms. Brooks
discussed various features of atmospheric electricity that vary with solar activity and appear to be related to cosmic ray ionization. Especially intriguing is his suggestion that known correlations between meteorological phenomena and lunar phase might involve a modulation of cosmic ray fluxes by the moon disturbing the tail of the earth's magnetosphere. There is a hazard, however, in attempting to connect measurements of thunder and lightning to other meteorological processes. That is, the lightning frequency might vary with electric fields and ion concentrations but without other features of convective storms being significantly influenced. There have been numerous suggestions in the past that cloud electric fields could increase the rates of ice nucleation or cloud droplet coalescence, but the possible significance of such effects is still unclear (cf., e.g., Doolittle and Vali, 1975; Latham, 1969). On the other hand, Levin and Ziv (1974) have developed a thundercloud model that indicates electric fields play an important role in the levitation of raindrops. In any case, there are no models of electrified clouds sufficiently sophisticated to evaluate the possible role of changes in atmospheric ion content.

Even if the contribution of extratropical thunderstorms to the atmospheric heat budget (through latent heat release and cloud production) did vary with the solar cycle, it is not obvious that this variation would be reflected in significant climatic variations. No detailed evaluation of the contribution of extratropical thunderstorms to the global heat budget is available but it would seem that outside of the tropics this contribution must be rather small compared to the solar constant. The possible connection between atmospheric electricity, thunderstorms, and other meteorological processes merits further consideration, but I think other mechanisms may be more promising.

Variations of stratospheric ozone distribution with solar activity have been discussed by Ruderman and Chamberlain (1975) as regards galactic cosmic rays, and by Crutzen et al. (1975) as regards solar protons. Such variations will provide some modulation of tropospheric radiation balance, but this modulation appears to be too small to be significant. I shall now discuss what seems to me the most likely connection of cosmic ray ionization to weather. Roberts and Olson (1973) speculate that "incoming solar particle streams associated with geomagnetic disturbances and auroras cause increased ionization at or near the 300-mb level, which in turn leads to formation of cirrus near the tropopause." It is indeed known that ions nucleate water droplets from vapor phase at significantly lower supersaturations than the vapor pressures at which homogeneous nucleation will occur (Byers, 1965). However, at least several hundred percent supersaturation would still be necessary. Such high values of supersaturation never occur in the troposphere because there is always a sufficient number of particles to provide sites for heterogeneous nucleation at vapor concentrations not much greater than 100%. Furthermore, the frequency at which solar particle streams reach the tropopause is apparently rather low.

With some modifications, however, the Roberts' hypothesis can be refurbished to provide a somewhat more plausible model. First, as we argued at length in the previous section, the dominant source of ionization in the troposphere and lower stratosphere is galactic cosmic rays. If cloud formation is to be related to ionization, it must involve these galactic cosmic rays. Furthermore, in high latitudes at tropopause level and above, the fractional variation of galactic cosmic ray ionization with solar activity is significant compared to the mean ionization rates.

I shall now argue that whereas it is unlikely that ionization directly nucleates cloud droplets, it still could be very important for stratospheric aerosol formation, and this aerosol in turn could easily nucleate cloud droplets. The major component of stratospheric aerosol is a sulfuric acid-water mixture, possibly also containing some nitric acid (e.g., Kiang and Hamill, 1974). Large concentrations of aerosol are found down as low as 12 km in polar latitudes but primarily above 18 km in equatorial latitudes (Lazrus and Gandrud, 1974). The sulfuric acid component is produced as gas from sulfur dioxide. The latter is apparently oxidized primarily by HO and HO compounds (Davis, 1974). The ~70% sulfuric acid component of the aerosol greatly lowers the saturation vapor pressure of water over its pure value, and ~10% nitric acid may provide further vapor pressure depression. An analysis by Kiang and Cadle (1975) shows that, within the range of conditions found in the lower stratosphere, the ambient sulfuric acid in conjunction with the appropriate fraction of water vapor may either diffuse almost entirely to existing particles, or may largely form new particles by homogeneous nucleation. Kiang and Cadle do not discuss the origin of the existing particles. The relevant question concerning these is whether they were originally nucleated in the stratosphere or were carried up as solid particles from the troposphere. A distinction should be made between new particle formation as a sink for sulfuric acid and as a source of new particles. It is likely that the concentrations of sulfuric acid necessary for most new nuclei to form in the stratosphere are considerably less than those required for the bulk of the sulfuric acid molecules to go into new particles. The aerosol mass loss to the troposphere by hydrodynamic transport and sedimentation presumably involves primarily particles with radii of several tenths of a micrometer or greater. Most of the stratospheric particles are observed to be in this size range. Since embryonic nuclei just large enough to grow by diffusion have radii ~5·10^4 µm, all the required new nuclei could be formed in situ using less than 10^6 of the available sulfuric acid molecules.

It appears likely to me that aerosols will nucleate
preferentially on existing clusters rather than homogeneously. The question of nucleation in both cases involves the random formation of molecular aggregates of critical size such that further additions to molecules will lower the free energy of the system. This generally requires a cluster of several tens of molecules. The free energy per molecule decreases with decrease in size of clusters less than critical so that formation of critical clusters is rare until sufficiently high mixing ratios of the nucleating gases (here H\(_2\)O and H\(_2\)SO\(_4\)) are present. A cluster forming randomly about an ion has a lower free energy barrier to overcome. Indeed, small subcritical clusters will form that are stable, that is they are of lower energy than the ion itself on account of electrostatic binding forces. The most stable such positive ion aggregate possible in the atmosphere is of the form H\(_n\)O\(^+\)(H\(_2\)O)\(_n\), where \(n\) is between 5 and 7 (Mohnen, 1971; Castleman, 1974). Any positive ion that is created will inevitably go through a series of exchange reactions until it reaches this form. Thus the positive ions in the stratosphere (which have concentrations \(\sim10^{6}\) cm\(^{-3}\)) must consist largely of such clusters.

The question raised here is whether critical sized nuclei will form on these ions at a rate greater than their rate of homogeneous nucleation. If so, the rate of aerosol nuclei formation will be proportional to that of ion concentration. With decreases of ion concentration, fewer particles will form, and these, assuming no change in the rate of H\(_2\)SO\(_4\) formation, will therefore grow more rapidly and reach larger sizes than they would have otherwise.

To finish my speculative chain, I must link stratospheric aerosol to cloud formation. First, however, I wish to point out that the division between stratospheric and upper tropospheric aerosols is not clearcut. One distinction is that particles in the troposphere have a residence time of at most months compared to years for the stratospheric aerosol, but even as regards residence times no sharp discontinuity at the tropopause is expected. Sulfate aerosol particles can presumably form just below as well as above the tropopause by the same mechanisms. In the lower troposphere, however, another mechanism involving oxidation of SO\(_2\) in cloud droplets is believed dominant (cf., Easter and Hobbs, 1974).

Sulfuric acid aerosol particles of a few tenths of a micrometer in radii and mass of around \(10^{-13}\) g represent very efficient cloud condensation nuclei. Their solubility in water greatly depresses the minimum vapor pressure that would be necessary for growth of a cloud drop compared to that needed for homogeneous nucleation or even on an insoluble particle of the same size (cf., e.g., Byers, 1965, Fig. 2.4).

The crux now of my speculative model is the assumption that sulfuric acid aerosol formed either just above or just below the tropopause:

a) has properties that vary with fluctuations in ion concentrations;

b) is the dominant nucleating agent of clouds at these levels (at least in middle and high latitudes).

At this point, I have so piled speculation upon speculation that much further argument does not seem profitable. In particular, I will not say whether the creation of fewer but larger aerosol particles at times of maximum solar activity (and minimum cosmic ray incidence) should increase or decrease cloudiness. The prediction of the sign of this effect is surely more difficult than that of cloud seeding where insertion of a certain level of cloud nuclei can enhance the growth of rain drops in a cloud but larger concentrations of nuclei can dissipate the cloud.

Finally, possible implications of the observed large variations of stratospheric aerosol with volcanic activity should be mentioned. First, these volcanically induced variations will make it more difficult to identify fluctuations related to solar activity. Second, if this aerosol modulates cloud formation, volcanic activity may be more important for climate than heretofore believed.

5. Concluding remarks

I hope that this discussion has provided some guidance as to fruitful avenues for further research into physical connections between solar activity and the lower atmosphere. For those skeptics (myself included) who find it difficult to accept the likelihood of such connections, these arguments point out several important meteorological processes, about which we know so little that it is not difficult to use them to build speculative links between solar activity and weather. These include, in particular, the role of ions in the formation of sulfuric acid aerosol in the lower stratosphere, and the role of sulfuric acid aerosol in nucleating clouds in the vicinity of the tropopause.

References


Mohnen, V. A., 1971: Discussion of the formation of major positive and negative ions up to the 50 km level. Pure Appl. Geophys., 84, 141–153.


**Announcements**

**NCAR postdoctoral and UCAR graduate fellowships**

The National Center for Atmospheric Research has issued announcements inviting applications for the 1976-77 Advanced Study Program’s Postdoctoral and Senior Postdoctoral Fellowships and for the 1976-77 UCAR Fellowship Program for graduate study leading to the Ph.D. in atmospheric sciences or related disciplines. The deadline for Postdoctoral and Senior Postdoctoral Fellowship applications is 1 February 1976; for UCAR Fellowships, 15 January 1976. For information contact: Dr. Jackson Herring, Acting Chairman, Advanced Study Program, NCAR, P.O. Box 3000, Boulder, Colo. 80305.

**AMS publication—Workshop on meteorology and environmental assessment**

Publication of Workshop on Meteorology and Environmental Assessment ("Lectures on Air Pollution and Environmental Impact Analyses") is expected by the end of January. The book will be available from the American Meteorological Society in hardbound copies only at $20 members; $25 nonmembers. All orders received prior to 30 January will be honored at the prices listed under Conference Preprints/Proceedings on 1976 dues and subscription renewal forms.

**NSF fellowships for science applied to societal problems**

The National Science Foundation (NSF) has reopened its annual competition for Faculty Fellowships in Science Applied to Societal Problems. Awards will be offered in all fields of science to science teachers at two- and four-year colleges and universities. The application deadline is 6 February 1976; the awards will be made in mid-April. For information and application materials contact: Faculty Fellowships in Science Applied to Societal Problems Program, Division of Higher Education in Science, National Science Foundation, Washington, D.C. 20550 (Tel: 202 282-7555).