absorption, which includes Doppler broadening and collision broadening, is expected to have this width. The H_2O concentration is $\sim 7.5 \times 10^{11}$ molecule cm^{-3} [VMR $\sim$ 1.5 parts per million (ppm)], indicating a very dry stratosphere (*11*) at 28 km since the saturated H_2O concentration at the ambient temperature of 225 K is $\sim 1 \times 10^{15}$ molecule cm^{-3}. Data from H_2O runs at other times have yet to be analyzed (*7*).

In summary, we report a new and accurate technique for measuring and analyzing trace constituents in the stratosphere. Although thus far we have measured only NO and H_2O, the technique is applicable to real-time measurements of other constituents important in stratospheric chemistry. The measurements carried out as a function of time show the influence of sunlight, in addition to giving the first measurements of NO concentration at 28 km. The measured daytime value of $\sim 2 \times 10^9$ molecule cm^{-3} and the nighttime value of $\lesssim 1.5 \times 10^8$ molecule cm^{-3} may allow us for the first time to put some of the models of stratospheric chemistry and the O_3 cycle on a much more concrete footing. It is also important to investigate the variation of NO as the sun sets, thereby removing the NO generation source. The sunset measurements are eminently possible with this new technique and will show the recombination effects of NO and O_3 which are important in the O_3 cycle. These measurements, together with simultaneous measurements of O_3 concentrations are planned for the future.

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12. We would like to acknowledge the tremendous help given by V. K. Smith, F. Crutcher, R. Myers, and their colleagues (division 1255) at Sandia Laboratories without whose collaboration the experiment reported here could not have been carried out. Thanks are also due to Dr. G. L. Miller of Bell Telephone Laboratories for the design of the ultralow-noise preamplifiers used with the OA cell, to R. J. Kerl for assistance in the early parts of the experiments and in the construction of the OA cells, to J. H. Hasiak and C. Pace for assistance in the mechanical construction of the experiment, and to C. Yuen (Swarthmore College) who tested and calibrated the OA cells. In addition, we thank R. H. Rabiner (Data General Corporation) whose assistance with the minicomputer used in this experiment was indispensable.

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Acid Rain: A Serious Regional Environmental Problem

Abstract. *At present, acid rain or snow is falling on most of the northeastern United States. The annual acidity value averages about* p*H 4, but values between* p*H 2.1 and 5 have been recorded for individual storms. The acidity of precipitation in this region apparently increased about 20 years ago, and the increase may have been associated with the augmented use of natural gas and with the installation of particle-removal devices in tall smokestacks. Only some of the ecological and economic effects of this widespread introduction of strong acids into natural systems are known at present, but clearly they must be considered in proposals for new energy sources and in the development of air quality emission standards.*

As part of the Hubbard Brook Ecosystem Study, we have monitored the chemistry of precipitation in north-central New Hampshire for about 11 years (*1*) and have found surprising acidity (*2*). Normally water in the atmosphere in equilibrium with prevailing CO_2 pressures will produce a *p*H of about 5.7 (*3*), but much stronger acids have recently been observed in rain and snow in the northeastern United States, with *p*H values as low as 2.1. The presence of these acids is presumably related to air pollution (*2*).

Current measurements and the few scattered observations during the past 11 years show that precipitation falling in northeastern United States is significantly more acidic than elsewhere in the United States (*2, 4*). For example, the annual mean *p*H, based upon samples collected weekly during 1970–1971 and weighted proportionally to the amount of water and *p*H during each period of precipitation, was 4.03 at the Hubbard Brook Experimental Forest, New Hampshire; 3.98 at Ithaca, New York; 3.91 at Aurora, New York; and 4.02 at Geneva, New York. Measurements on individual rainstorms frequently showed values between *p*H 3 and 4 at all of these locations. Data from the National Center for Atmospheric Research included precipitation *p*H values as low as 2.1 in the northeastern United States during November 1964 (*4*). Summer rains are generally more acidic than winter precipitation (*5*).

The major cation is H^+, which accounts for 44 and 69 percent of the cations (milliequivalent basis) in the Ithaca and Hubbard Brook precipitation, respectively. Of the anions in Ithaca and Hubbard Brook precipitation, respectively, $SO_4{}^{2-}$ represents 59 and 62 percent, $NO_3{}^-$ contributes 21 and 23 percent, and Cl^- constitutes 20 and 14 percent.

Precipitation *p*H values from the northeastern United States are similar to those recorded over southern Sweden. More than 70 percent of the sulfur in the air in Sweden is thought to be anthropogenic, of which 77 percent has its source outside Sweden (*6*). The mean residence time for sulfur in the atmosphere is estimated to be 2 to 4 days (*7*). As a result, SO_2 may be transported more than 1000 km in this region before being deposited. Distant industralized regions, such as England and the Ruhr Valley, are thought to be major contributors to Sweden's acid rain (*8*). Moreover, Odén's data show a striking increase in precipitation acidity in northeastern Europe over the past two decades, with the H^+ concentration of rain in some parts of Scandinavia increasing more than 200-fold since 1956 (*9*).

We know of no long-term record of

precipitation acidity for the United States. Recent data, since 1963, do not reveal any marked trends (Fig. 1), although extensive data on precipitation chemistry from Geneva, New York, provide indirect evidence of higher *p*H's in earlier years (*10*).

Prior to 1940 *p*H was not measured, but methyl orange was used to indicate acidic or alkaline conditions in some of the samples collected in New York State. Since methyl orange changes color at *p*H 4.6, little exact information was obtained; but, significantly, no acidic reactions were recorded. In addition, Collison and Mensching (*11*) found relativity large amounts of HCO_3^- in precipitation at Geneva prior to 1930 (*12*). Because HCO_3^- does not coexist with stronger acids such as those found in today's rain, these data are consistent with the hypothesis that acid rain is a recent phenomenon in the northeastern United States.

Data on the sulfur content of rain and snow in New York State indicate that present-day precipitation contains about 70 percent less sulfur than that prior to 1950 (Fig. 2) (*13*). This drop in sulfur concentration at both Ithaca and Geneva is difficult to reconcile with the proposed recent increase in acidity. High concentrations of sulfur in precipitation could reflect the presence of H_2SO_4. Since the *p*H of the precipitation apparently was not lowered prior to 1950, much of the sulfur must have existed as un-ionized particulate sulfate or as ionized forms neutralized by equivalent amounts of bases (*14*). Gorham (*15*) observed large amounts of calcium in smoke solids collected from chimneys in London and argued that such basic substances were more easily and quickly precipitated than acidic ones. His results and those of Overrein (*16*) indicated that various basic particles in smoke would neutralize acids and tend to fall out relatively near the pollution source. Overrein also found the lowest *p*H's some distance downwind from the combustion source, an indication that a surplus of acid-forming substances escaped neutralization and gave rise to acid rain (*16*).

It appears that much of the sulfur in early precipitation (before 1950) at Ithaca and Geneva resulted from the local combustion of coal (*5*). Prior to 1932, coal and wood were used almost exclusively as sources of fuel in Ithaca, although some manufactured coal gas was used. Natural gas was introduced into the Ithaca area late in 1932 but did not really replace coal as the prime fuel until 1950–1955 (*17*). The difference in the sulfur content between coal (high) and natural gas (low) is enormous, and thus this shift in use patterns is highly correlated with the decline in the sulfur content of precipitation in 1950–1955 (Fig. 2). The effect of coal burning on local precipitation chemistry also is indicated by a shift in seasonal characteristics. Before 1950 the wintertime concentrations of SO_4^{2-} in precipitation always exceeded the summer values (*11, 12*), whereas now maximum SO_4^{2-} concentrations occur during the summer (*5*).

Significantly, appreciable quantities of NO_x are normally produced in the combustion of natural gas. The relatively large increase in the NO_3^- content of precipitation since about 1945 (Figs. 1 and 2) may be correlated with the increased combustion of natural gas and motor fuels (*5*).

We suggest that the change in acidity occurred soon after 1950 because of a shift in the predominant form of sulfur in the atmosphere, and even though there was a decrease in the total sulfur content of the precipitation. When the major source of anthropogenic sulfur for the atmosphere was the combustion of coal, much of the sulfur was precipitated to the land near the combustion source in particulate form and as neutralized salts. Today, with the increasing combustion of fossil fuels, with mounting numbers of taller smokestacks fitted with precipitators to remove the larger particles (*18*), and with increasing combustion of fossil fuels other than coal, greatly increased quantities of SO_2 apparently are being introduced into the atmosphere, at least on a regional basis (*19*). Injected at heights of 60 to 360 m, the SO_2 may be dispersed over wide areas; and, in the absence of equivalent amounts of alkaline substances in the atmosphere, appreciable quantities of SO_2 are converted to acid. The consistently low *p*H of precipitation at rural sites in New England (*2, 4*), hundreds of kilometers from urban industrial centers, attests to the long-range dispersion of SO_2 and its secondary pollutant, H_2SO_4. It would appear, then, that these trends in fuel consumption, fuel preference, and pollution control technology (increasing the height of smokestacks and installing particle precipitators) have transformed local "soot problems" into a regional "acid rain problem."

The ecological effects of acid rain are as yet largely unknown, but potentially they are manifold and very complex. Effects may include changes in the leaching rates of nutrients from plant

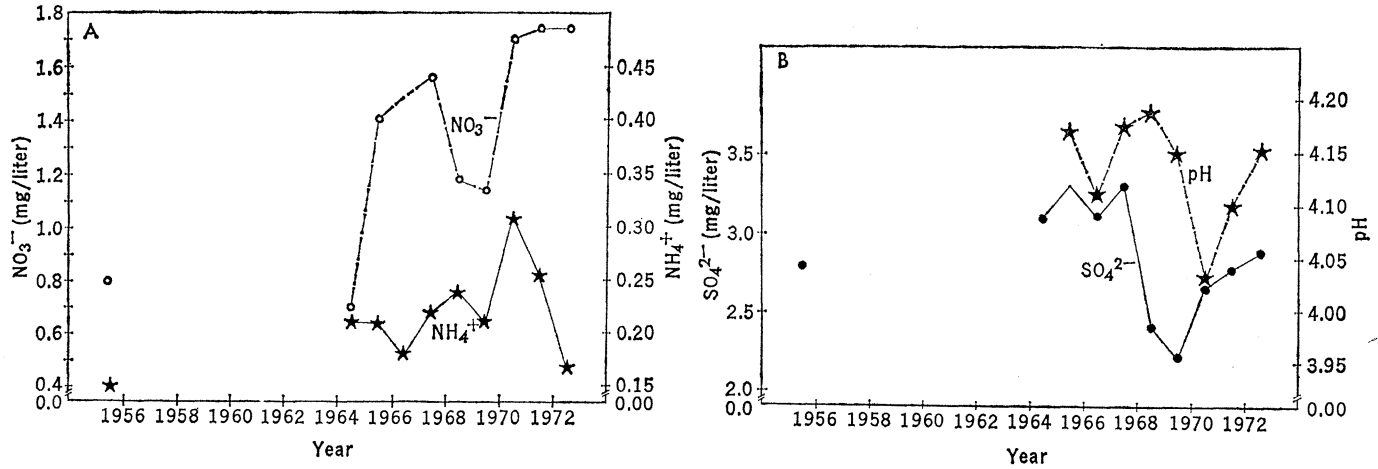

Fig. 1. Weighted annual concentrations of NO_3^- and NH_4^+ (A) and SO_4^{2-} and the *p*H (B) of precipitation at the Hubbard Brook Experimental Forest in New Hampshire. Values for 1955–1956 were estimated from Junge and Werby (*35*) and Junge (*36*).

foliage, changes in the leaching rates of soil nutrients, effects on predator-prey relationships, acidification of lakes and rivers, effects on the metabolism of organisms, and the corrosion of structures.

It is believed that a reduction in forest growth in northern New England and in Scandinavia over the last two decades may be correlated with the concurrent acidification of precipitation (*8, 20*). Study of the direct effects of acid rain on plants is just beginning. Gordon (*21*) has observed that the emerging needles of several species of western pines, when inoculated with an atomized solution of H_2SO_4 at a $pH < 4.0$, grew to only half the length of control needles. Wood and Bormann (*22*) noted spot necrosis and irregular development of leaf tissue on deciduous tree seedlings (yellow birch) 2 to 5 weeks old subjected to misting with aqueous H_2SO_4 at pH 3.0. The 2-week-old seedlings were more susceptible than the 5-week-old plants. In studies of tomatoes on the island of Hawaii, rain of low pH was found to decrease pollen germination and pollen tube growth and generally lower the quality and production of these plants (*23*). Toxic effects on plants may be associated with the increased proportions of HSO_3^- ions and undissociated H_2SO_3 at low pH (*24*).

Of equal concern are potentially numerous indirect effects (direct nutrient leaching or the erosion of the cuticle, providing ready access to pathogens, herbivores, or nutrient leaching) whereby the general vigor of plants may be reduced by acid precipitation and synergistically contribute with other impinging stresses to an adverse effect. In deciduous forests of the northeastern United States, the precipitation is generally most acidic during the growing season (*25*).

Increased input of acid to soil may lead to increased leaching of calcium and other nutrient elements [for example, see (*8, 16*)]. Although such losses would be small for some ecosystems and unlikely to result in any significant short-term damage to arable land, they do represent other added stresses to the ecosystem. During the growing season in forested areas, most of the acid rain impinges directly on the foliage where ion exchange occurs. Thus the rain that reaches the forest floor (soil) is far less acid than the incident precipitation (*26*).

The effect of acid precipitation on aquatic ecosystems may be large, particularly if the input ratio of direct precipitation to land drainage is high. A significant proportion of the decreasing trend in pH, observed during 1965–1970 in a large number of lakes and rivers in Scandinavia, has been attributed to acid rain (*27*). In Canada, numerous lakes west of the Sudbury smelters have increased in acidity by more than 100-fold during the last decade (*28*); by 1970 some 33 of these lakes had pH values less than 4.5 and were termed "critically acid." Schofield (*29*) reports that the water chemistry of a relatively large clear-water, oligotrophic lake with a small drainage area in the Adirondack Mountains of New York has changed appreciably since 1938. In December 1938 the total alkalinity, expressed as $CaCO_3$, ranged from 12.5 to 20.0 mg/liter and the pH was 6.6 to 7.2, whereas during 1959–1960 the alkalinity ranged from 0 to 3.0 mg/liter and the pH was 3.9 to 5.8.

Serious fish mortality, particularly early age classes in salmonids, has been reported in Scandinavian rivers and lakes (*8, 30*) and Canadian lakes (*28*) and has been attributed directly to increased acidity from precipitation. Similar acidification of aquatic ecosystems must be occurring in other parts of the world, but it is unrecognized or is often confounded by other sources of pollution [for example, see (*31*)].

Damage to buildings, structures, and art forms by acid precipitation may be enormous (*8, 32*). The economic consequences of this damage are only now becoming apparent.

An estimate of total anthropogenic emissions for the United States indicates that this country is now annually injecting about 32×10^6 metric tons of SO_x into the atmosphere (*33*). Additional projections suggest that this amount will increase two- to fivefold by the year 2000 (*33, 34*). Development of nuclear-based power, of methods for extracting "clean" energy from coal and oil, and the passage of more stringent regulations governing SO_x emissions may slow this trend. In general, though, it seems safe to assume that, as long as energy demands mount and fossil fuels remain abundant and economically desirable, SO_x emissions will increase. Surely this will be true if air pollution standards are relaxed to meet energy

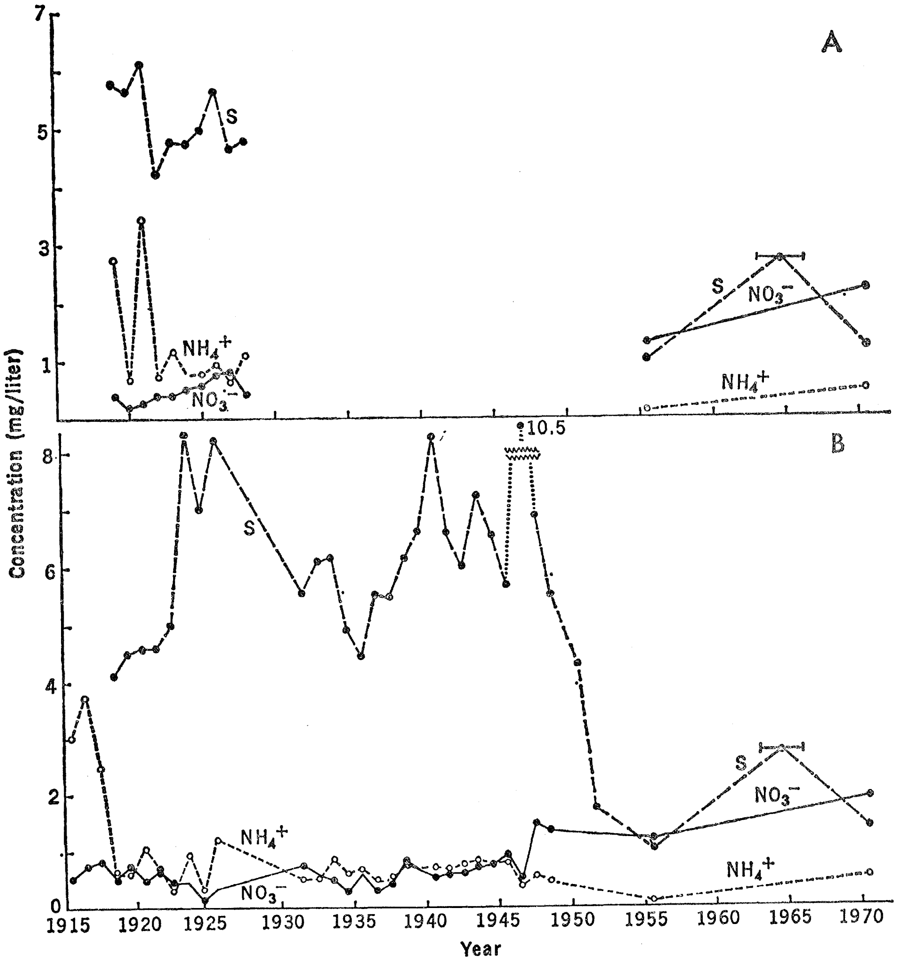

Fig. 2. Weighted annual concentrations of sulfur ($SO_4^{2-} - S$), NH_4^+, and NO_3^- in precipitation at Geneva, New York (A), and Ithaca, New York (B).

needs during the "energy crisis." The extent to which the SO_2 concentration may ultimately rise is a matter for serious concern, for increased concentrations may promote still more serious and widespread acid rain with all that this outcome implies for the structure and function of both natural and man-manipulated ecosystems.

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11. R. C. Collison and J. E. Mensching, *N.Y. Exp. Stn. Geneva Tech. Bull. No. 193* (1932): "Bicarbonates have been found in considerable abundance in all samples of water collected at Geneva and vary from 12 to 92 pounds as HCO_3^- per acre annually" (p. 4); "at this station (Table 5) bicarbonates (HCO_3^-) were present in all samples of rainwater analyzed both yearly and monthly. . . . More bicarbonate was present during the summer months than in the winter months. As already noted all samples of rainwater collected contained this constituent" (p. 16). In the absence of data on *p*H, the large amounts of HCO_3^- in early rainwater samples at Geneva, New York (1919–1929) indicate much higher *p*H's than today. Indicator solutions (methyl orange and phenophthalein) were used to measure concentrations of HCO_3^- and CO_3^{2-} in the precipitation (p. 10), hence the above statements would clearly indicate that all samples would have had a $pH > 4.6$. Only trace amounts of HCO_3^- are found in present-day precipitation samples at Geneva. The presence of HCO_3^-, therefore, would indicate that *p*H values in 1919–1929 were probably 5.7 or higher.
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13. Data for Geneva and Ithaca are derived from various published sources; procedures for collection and analysis of samples have been evaluated in relation to observed trends in precipitation chemistry (*5*). Wilson (*12*) believed that, prior to 1923, the area adjacent to the precipitation collector at Ithaca was relatively free of smoke. However, in 1923 a large heating plant (Cornell University) was constructed within 1.6 km from the collection area and the sulfur content of precipitation (annual averages) was thereafter appreciably increased, at times more than doubled. At Geneva a branch railroad passed within 183 to 214 m of the precipitation collector and a coal-burning flour mill was also nearby (*11*). Various events, including the economic depression of the 1930's, the World War II industrial effort in the early 1940's, and the coal miners' strike in 1945, have undoubtedly affected the amounts of nitrogen and sulfur in precipitation.
14. An alkaline response to methyl orange indicator was observed in every precipitation sample collected in Tennessee during 1919–1921 in spite of high concentrations of sulfur in the samples [W. H. MacIntire and J. B. Young, *Soil Sci.* **15**, 205 (1923)]. This was attributed to excess amounts of "lime" and other neutralizing bases in soot and fine dust found in precipitation samples. Moreover, the total amount of sulfur precipitated was about five times greater than the amount in water-soluble forms and was directly related to the amount of solid soot falling directly or with precipitation.

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A Greater Gondwanaland

Abstract. *Gondwanaland at its maximum extent included Tibet, the Tarim Basin block, and parts of northern China. Tibet lay between India and Western Australia as submerged continental crust. The Himalayas are of intracontinental origin. The Tien Shan lie along the former oceanic boundary of Gondwanaland.*

In Gondwanaland reassemblies, two important problems are the present location of the continental crust which lay between Australia and India (*1*) and the present relation of India to the rest of Asia. The Himalayas are believed (*2*) by some to be the product of continental collision preceded by subduction of Tethyan oceanic crust. Critics of continental drift (*3*) have stressed that there are enigmatic relationships between India and the rest of Asia which are inconsistent with such collision.

I suggest that the two problems are closely related. Gondwanaland was much larger than generally believed; it included Tibet and the Tarim Basin block, the Himalayas having formed intracontinentally. Evidence against collision comes from the anomalous position of the Indus Suture Line (*4*) which, if it is the relic of oceanic subduction preceding collision, lies on the wrong side of the Himalayas. I have suggested (*5*) that the line is a relic of a Permojurassic oceanic opening to the