strike responses for 132 hours after conditioning (P < .01; Mann-Whitney U test). The threshold of the bite-strike response of the experimental animals, however, continued to be significantly different from the threshold of the controls for over 180 hours (Fig. 2F; P < .01 through 156 hours, P < .02 at 180 hours; Mann-Whitney U test). After 180 hours the observations on extinction were terminated so that the animals could be reconditioned in other experiments (8).

The results of this investigation demonstrate that pairing or contiguity of food (CS) and shock (UCS) are necessary to produce the long-term changes in the withdrawal and feeding behavior of the experimental animals. As further required in classical conditioning (5, 7), experiments designed to control for pseudoconditioning and sensitization, in which food and shock were presented unpaired to the control animals, did not yield the same long-term behavioral changes as were produced by paired stimulation. We interpret these data to show that the rapid and long-lasting behavioral changes of the experimental animals are attributable to associative learning.

The ability of the present paradigm to generate rapid learning of food aversion is striking, particularly when one considers that no alternative, safe food was made available and that the animals were maintained in a relatively starved condition. The drive to feed is usually so strong in Pleurobranchaea that feeding thresholds remain stable even after electroconvulsive shocks (4). It would be expected, therefore, that food stimuli would be strong reinforcers of behavior and that hungry animals would be difficult to train to avoid food. Previous experiments on Pleurobranchaea show, however, that a paradigm which used a food stimulus to reinforce conditioned feeding responses to mild tactile stimulation was much less effective than the food aversion paradigm we used here

This apparent contradiction may be placed into perspective by viewing the stimulus-response repertoire of Pleurobranchaea in the context of behavioral hierarchy which describes how an animal will behave when confronted simultaneously by stimuli that elicit different responses (4). This hierarchy may also be viewed as a representation of the drive or motivation-related value of different stimuli with respect to the total behavior of the animal. The hierarchy of three of the behaviors of Pleurobranchaea in increasing dominance, and the inferred value of the associated stimuli in increasing strength, is: withdrawal, elicited by mild tactile stimulation; feeding, elicited by food; and

escape, elicited by strong noxious stimulation. The results of the learning studies on Pleurobranchaea are consistent with the notion that the reinforcing power of a stimulus is governed by its drive or motivational value (7); tactile stimulation has been used only as a CS, food as a CS or UCS, and noxious stimulation only as a UCS. As a first approximation, we can say that the lower the position of a stimulus on the hierarchy the greater will be its utility as a CS, while the higher its position the greater will be its effectiveness as a UCSas a reinforcer. Thus, in the context of the behavioral hierarchy of Pleurobranchaea, it is not unexpected that strong noxious stimulation would be a more effective reinforcer than food.

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References and Notes

1. E. R. Kandel, in The Neurosciences: A Study Program, G. C. Quarton, T. Melnechuk, F. O. Schmitt, Eds. (Rockefeller Univ. Press, New York, 1967), vol. 2, pp. 666–689; F. B. Krasne, in *Invertebrate Learning*, W. C. Corning, J. A. Dyal, A. O. D. Willows, Eds. (Plenum, New York, 1973), pp. 49–130; A. O. D. Willows, in *ibid.*, pp. 187–

- 274; J. V. McConnell, Annu. Rev. Physiol. 28, 107
- (1966). G. J. Mpitsos and W. J. Davis, *Science* **180**, 317
- (1973). W. J. Davis and G. J. Mpitsos, Z. Vgl. Physiol. 75,
- W. J. Davis and G. J. Mpitsos, Z. Vgl. Physiol. 75, 207 (1971); W. J. Davis, M. V. S. Siegler, G. J. Mpitsos, J. Neurophysiol. 36, 258 (1973); M. V. S. Siegler, G. J. Mpitsos, W. J. Davis, ibid., in press. W. J. Davis, G. J. Mpitsos, J. M. Pinneo, J. Comp. Physiol. 90, 207 (1974); ibid., p. 225.

 1. Gormezano, in Experimental Methods and Instrumentation in Psychology, J. B. Sidowski, Ed. (McGraw-Hill, New York, 1966), pp. 385-420; and J. W. Moore, in Learning Processes, M. H. Marx, Ed. (Macmillan, Toronto, 1969), pp. 121-203.
- Parametric tests were used to analyze withdrawal data. Variance was calculated with the chi-square (χ^2) test. Confidence limits on pooled withdrawal data were determined for a proportion with un-known but true standard deviation as per G. W. Snedecor and W. G. Cochran, Statistical Methods (Iowa Univ. Press, Ames, 1967), p. 593. The data on threshold and latency of the feeding responses were analyzed with nonparametric tests. Confidence limits were expressed as a function of standard error. The Mann-Whitney U test was standard error. The Mann-Whitney U test was performed for analysis of variance; for pooled data the sampling distribution of U rapidly approaches the normal distribution as per S. Siegel, Nonparametric Statistics for the Behavioral Sciences (McGraw-Hill, New York, 1956), p. 312.

 N. E. Miller, in The Neurosciences: A Study Program, G. C. Quarton, T. Melnechuk, F. O. Schmitt, Eds. (Rockefeller Univ. Press, New York, 1967), vol. 2, pp. 643–652.

 G. J. Mpitsos and S. D. Collins, in preparation. We thank N. Robbins, K. Alley, and A. Brennan for helpful criticisms on the manuscript; J. Phillips and B. Cunningham for assistance on the statisti-
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Acidity in Rainwater: Has an Explanation Been Presented?

The recent report by Likens and Bormann (1) certainly points out the manifold ecological problems, both present and potential, that are associated with the possibility of acid rain. It cannot be disputed that the tall smokestacks introduced in recent years disperse the emitted SO, over a broader area. That is precisely the philosophy behind tall stacks. Whether there has been a concomitant increase in acidity in the rainwater of rural areas in the northeastern United States has not been documented in the data of Likens and Bormann since, as shown in their figure 1, the pH remained relatively constant over an 8-year

The authors, however, do show in their figure 2 a striking 70 percent decrease in SO₄²- since 1950 in rainwater at Ithaca, New York. This change is probably correctly attributed to the shift in use from coal (high in sulfur) to natural gas (low in sulfur). The statement is made (correctly) that the drop in the sulfur concentration is difficult to reconcile with the proposed (not substantiated) recent increase in acidity. The contention is then made that high particulate loadings from the combustion of coal in the past could have caused neutralization of the acid sulfate. Likens and Bormann further conclude that the installation of particle-removing devices in tall smokestacks eliminates these alkaline substances, consequently permitting appreciable quantities of SO₂ to be converted to

This argument cannot be correct. As an example, coal could contain 3 percent sulfur (6 percent as SO₂) and 15 percent ash (composed of 50 percent SiO₂, 30 percent Al₂O₃, 15 percent Fe₂O₃, and 3 percent CaO). The alumina and silica are quite inert, and there is just not enough material left to neutralize the SO₂. The only component of any possible significance is Fe₂O₃. Even if we consider the highly unlikely overall reaction

$$3 SO_2 + Fe_2O_3 + \frac{3}{2} O_2 \rightarrow Fe_2(SO_4)_3$$

it would require essentially all of the Fe₂O₃ to consume an appreciable amount of the SO2. Since there are so many competing processes, it is most unreasonable to assume that a reaction could appreciably proceed when the reactants are present in a near stoichiometric relationship. Furthermore, 25 years ago particulate-removal devices in operation in the northeast, if they were mechanical, removed > 50 percent of the ash or, if electrostatic (in common usage), > 75 percent. Modern electrostatic precipitators remove ~ 99 percent of the ash. This increase in particulate removal made possible by the use of modern electrostatic precipitators is hardly enough to be significant for the enormous difference in neutralization proposed by the authors. Finally, there is no evidence that $Fe_2(SO_4)_3$ was ever a major component of rainwater SO₄²⁻.

Certainly, the introduction of tall smokestacks and particulate-removal devices have reduced the local "soot problem." Whether these procedures have altered (positively or negatively) the regional "acid problem" is an unanswered question.

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References

1. G. E. Likens and F. H. Bormann, Science 184, 1176 (1974).

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Both Newman and the news media have chosen to emphasize a speculative conclusion from our report (1) rather than the critical points that acid precipitation is a by-product of the combustion of fossil fuels, is falling over most of the eastern United States, and may have potentially serious ecological effects.

It seems clear that this regional "problem" is a recent phenomenon in the eastern United States, since (i) bulk precipitation samples collected during the first half of this century in the eastern United States contained significant amounts of HCO₃ and were not as acid as samples collected since 1950 (2, 3); (ii) acid precipitation has been falling on most of the northeastern United States since at least 1955-1956, yet both the intensity and particularly the area of acid deposition (pH < 5.6) have increased markedly since then (3); (iii) records from the Hubbard Brook Experimental Forest in New Hampshire show that, although the concentration was variable [figure 1 in (1)], the annual H⁺ input

in precipitation increased by 36 percent during the 10-year period from 1964–1965 through 1973-1974 (4); (iv) based on a stoichiometric formation process, the neutralizing component (5) in precipitation samples from central New York State was much lower after 1950 than before, and at Hubbard Brook the neutralizing component decreased from 40 µeq/liter in 1955-1956 to 25 μ eq/liter in 1965-1966 and to 18 μ eq/liter by 1973–1974, even though in the 1955-1956 and 1965-1966 samples dry fallout was excluded and in the 1973-1974 samples dry fallout was not excluded (3); and (v) although the SO₄2concentrations in precipitation samples from central New York State dropped during the mid-1950's, the NO₃ concentrations have increased since then (2), and the contribution of SO₄²⁻ to precipitation acidity (based on a stoichiometric balance) dropped from 78 to 65 percent between 1955 and 1973, whereas the contribution of NO; increased from 22 to 30 percent during the same period (3).

Whether particle removal from combustion gases has contributed significantly to the increased regional acidity of precipitation remains an unanswered question. However, the following points should be noted: (i) no other explanation has been put forth by Newman or others to explain the results of Gorham (6) and particularly Overrein (7), as cited in our report (1), which apparently show that particles in smoke are very effective in neutralizing acidity in local precipitation; (ii) likewise, no alternate explanation has been offered for the "substantiated" recent drop in SO₄²⁻ concentrations in central New York precipitation, coexisting with low pH values, whereas earlier high concentrations of SO₄² in precipitation samples coexisted with higher pH values [also substantiated (2, 3)]; and (iii) fly ash may be highly alkaline and produce high pH values in water suspensions (8). In view of the importance of aerosols and larger particles in neutralizing H⁺ in bulk precipitation samples, we would like to see some field or experimental data to support Newman's claim that particle removal may not permit "appreciable quantities of SO₂ to be converted to acid" (emphasis ours).

Important points that should not be overlooked are that the acid precipitation problem in the northeastern United States is due to increased amounts of strong acids (H₂SO₄ and HNO₃) in precipitation coincident with increased combustion of fossil fuels and to the regional dispersal of combustion gases from taller smokestacks.

Although we cannot say with certainty when precipitation in the northeast generally became acid (pH < 5.6), that is, whether it was 20 or 35 years ago, it seems that the major change may have been associated with the other dramatic changes in the precipitation chemistry of the mid-1950's (1). Although the precise date is elusive, it is clear that the pattern and intensity of acid deposition are recent and increasing, that the root cause is most likely the combustion of fossil fuels, and that this problem may be further exacerbated by a careless rush to solve the energy crisis.

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References and Notes

- 1. G. E. Likens and F. H. Bormann, Science 184,
- G. E. Likens and A. A. B. Likens and A. A. B. Likens, Cornell Univ. Water Resour. Mar. Sci. Cent. Tech. Rep. 50 (1972).
 C. V. Cogbill and G. E. Likens, Water Resour. Res. 10, 1133 (1975); C. V. Cogbill, thesis, Cornell University (1975). University (1975).
- University (1975).

 4. Based on a regression equation, $Y = 0.033 \ X + 0.83$, where Y is the H+ input in kilograms per hectare and X is the year. The correlation coefficient was .74 and the probability of a larger F value was .01.
- 5. By "neutralizing component" we mean all cations, scept H+, in excess of that contributed by sea salt
- 5). . Gorham, *Geochim. Cosmochim. Acta* 7, 231
- E. Goffman, Geochin. Cosmochin. Neta., 231 (1955).
 L. N. Overrein, Ambio 1, 145 (1972).
 D. G. Shannon and L. O. Fine, Environ. Sci. Technol. 8, 1026 (1974).

6 February 1975