Table 1. Nanograms of chlorinated hydrocarbon pesticides per liter of lake water associated with each fraction of particulates. Samples 79 and 75 show the qualitative presence or absence of chlorinated pesticides in each extracted fraction of two other samples taken at different times of the year.

Pesticide	Gradient fraction			
	1	2	3	4
		Sample 86		
Lindane	0.53	4.6	3.2	16.5
Heptachlor	0.69	0	1.7	2.1
Aldrin	14.7	2.0	1.0	1.3
Endrin	9.6	0	0	0
DDT: metabolic products or isomers	0	0	0	0
		Sample 79		
p,p′-DDD	+	+		+
Lindane	+		+	
Heptachlor	+	+	+	+
p,p′-DDT	+		+	
o,p′-DDT		+		
Endrin		+		+
o,p′-DDD			+	
		Sample 75		
o,p′-DDD	+	+	+	
p,p′-DDD	+	+	+	+
o,p′-DDT	+	+	+	+
p,p′-DDT		+	+	+
Lindane		+		+
Heptachlor		+	+	+
Endrin			+	

Pesticides were tentatively identified by comparison of retention times to those of control samples of known purity (dieldrin, aldrin, endrin, 99+ percent, Shell Chemical Co.; DDT and isomers, 99+ percent, USPHS pesticide repository; lindane, heptachlor, 99+ percent, City Chemical Corp.), and also by reinforcement of peaks by addition of known compounds. The distribution pattern of pesticides with different particulate fractions illustrates that various chlorinated hydrocarbons have an affinity for particulates separable by density-gradient centrifugation (that is, different particles). The distribution of these pesticides demonstrates their association with the particulate components suspended in the water. The utility of the collection and fractionation procedures is verified by the variety of pesticide separations obtained through its use.

Table 1 is a compilation of pesticide distribution and concentration in the particulate fractions of three separate lake water samples taken at different times, based upon GLC data without TLC confirmation, since the concentration of pesticide in gradient fractions is below the limit of TLC detection.

Evidence from this study suggests that there is a quantitative as well as qualitative distribution of pesticides associating with varied particulate components in natural waters. Pesticides that were determined were attached to particles of 0.15 μm or larger in size, and different pesticides were shown to be associated individually with particles of different density. For example, in sample 86, lindane was found in greater concentration in fraction 4, the inorganic portion of the particulate material (*5*), while aldrin and endrin were associated with less dense upper fractions 1, 2, and 3, which consist primarily of organics, detritus, and microorganisms. The presence of various chlorinated pesticides associated with different particulate components suggests a complex involvement of these compounds with components of the environment. These compounds could be present in the environment as either molecular aggregates in aqueous solution (4.1 nm or less) or in suspension (up to 0.11 μm) (*6*). In our system, particulates below 0.15 μm were not included, so that the presence of pesticides is indicative of some kind of true pesticide-particulate association. The particles could be cell, detritus, or inorganic materials (for example, clay minerals).

Different chlorinated hydrocarbons are present in different water samples. For example, samples 70 and 75 suggest the presence of DDT, its isomers, and metabolic breakdown products, whereas no DDT-group compounds were detected in sample 86. The lack of detection of DDT or isomers in this sample may be a reflection of the possibility of the specific association of such compounds with specific particulates, or, of course, it may be that these compounds were below the level of detection in the quantity of particulate used for the extraction.

In analyses where activated carbon filters have been used (for example, U.S. Public Health Service Water Pollution Surveillance Program) levels (0.05 ng to 0.05 μg/liter) of pesticides are detected. This technique has made use of 30-mesh high-grade carbon or layered 30-mesh and 4- by 10-mesh carbon which has been preextracted to remove organic material. It seems likely that chlorinated hydrocarbons associated with particles that have the general size of 0.15 μm could pass through coarse carbon filters and remain undetected in the water sample. Evidence from our study suggests that removal and analysis of particulates may need to be included to give more adequate estimates of pesticides in aquatic environments. In an environment such as Lake Erie, where the shallow water is so easily disturbed by wind action, the turnover and accumulation of pesticides in bottom sediments may be significant.

ROBERT M. PFISTER
PATRICK R. DUGAN
JAMES I. FREA
Microbial and Cellular Biology, Ohio State University, Columbus 43210

References and Notes

1. H. P. Burchfield and D. E. Johnson, *Guide to the Analysis of Pesticide Residues*, U.S. Dept. of Health, Education, and Welfare, Public Health Service Bureau of State Services (Environmental Health) (Government Printing Office, Washington, D.C., 1965).
2. L. Kahn and C. H. Wayman, *Anal. Chem.* **36**, 1340 (1964).
3. A. W. Breidenback, J. J. Lichtenberg, C. F. Henke, B. J. Smith, J. W. Eighelberger, Jr., H. Stierli, *The Identification and Measurement of Chlorinated Hydrocarbon Pesticides in Surface Waters*, U.S. Dept. of Interior, Federal Water Pollution Control Administration, 17-22 (Government Printing Office, Washington, D.C., 1966).
4. M. F. Kovacs, *J. Ass. Off. Agr. Chem.* **46**, 884 (1963); *ibid.* **49**, 365 (1966).
5. W. T. Lammers, *Limnol. Oceanogr.* **7**, 224 (1962).
6. M. C. Bowman, F. Acree, Jr., M. K. Corbett, *J. Agr. Food Chem.* **8**, 406 (1960).
7. Supported in part by grants A-006-OHIO and 013 Ohio, Office of Water Resources Research, U.S. Dept. of Interior. Mrs. Patricia Kennedy and Mrs. Virginia Price contributed technical assistance.

30 June 1969; revised 7 August 1969

Pulsar Test of a Variation of the Speed of Light with Frequency

Abstract. *The sharply defined optical and radio pulses from pulsars make possible a test of the variation of the speed of light with frequency, and of the possible existence of a photon mass. The data indicate that the mass of a real photon is less than 10^{-44} gram. Detection of extragalactic pulsars could allow a substantial improvement of this limit.*

The conventional description of light as a stream of massless particles implies that the speed of light in a vacuum is independent of its frequency. Direct tests of this independence have been made on earth by comparing the speed of radio waves with that of visible light (*1*). These speeds agree to about one part in 10^6. With the detection of the sharply defined radio and optical pulses from the pulsars, a substantial improvement of this comparison has become possible, both for visible light and for radio waves. This is done by comparing the arrival times of pulses at different frequencies, which presumably left the pulsar within one period of pulsation. An observed variation of arrival time with frequency for the radio waves has been attributed to the interaction with interstellar electrons. The size of this effect can, however, be used to place an upper limit on the intrinsic variation of velocity with frequency, provided that we assume that the two effects do not cancel. As will be shown below, in the case of a finite photon mass, the effect has the same sign as that of the electrons, and thus no cancellation can occur.

We note the relation between variation of velocity Δv and variation of arrival time Δt with frequency $\Delta\omega$

$$\frac{\Delta t}{\Delta\omega} = -\frac{l\,\Delta v}{c^2\,\Delta\omega} \quad (1)$$

where l is the distance to a pulsar and c is the speed of light. For the Crab Nebula pulsar NP 0532, optical pulses were detected in three colors within the same pulse (*2*). This implies that the variation of Δt across the visible region is less than, say, two pulse widths, or 3×10^{-3} second.

Hence in the visible region, say, for blue and violet light, we have (using $l = 2$ kiloparsecs)

$$\frac{\Delta v}{c} \lesssim \frac{c \times 3 \times 10^{-3}}{l} = \frac{0.9 \times 10^{8}}{6 \times 10^{3} \times 10^{18}} \lesssim 10^{-14} \quad (2)$$

which is many orders of magnitude better than terrestrial measurements. Even more precise limits for $\Delta v/c$ based on analysis of the Crab Nebula optical pulsar have recently been reported by Warner and Nather (*3*).

For the radio waves, the data are usually presented in terms of $\bar{n}_e$, the average density of electrons along the path. We have, from the usual plasma dispersion formula, in the approximation

$$(4\pi\bar{n}_e\,e^2)/m_e \equiv \omega_p^2 \ll \omega^2$$

where e is the charge of the electron, m_e is the mass of the electron, and ω_p is the plasma frequency,

$$\frac{\Delta v}{c} = \frac{2\pi\,\bar{n}_e\,e^2}{m_e}\left(\frac{1}{\omega_1^2} - \frac{1}{\omega_2^2}\right) \quad (3)$$

For the Crab Nebula pulsar $\bar{n}_e \lesssim 2.8 \times 10^{-2}$ electron/cm^3 from the observed dispersion (*4*). Hence

$$\frac{\Delta v}{c} \sim 1.6 \times 10^{9}\left(\frac{1}{\omega_1^2} - \frac{1}{\omega_2^2}\right)$$

or

$$\frac{\Delta v}{c} \lesssim 1.2 \times 10^{-9}$$

between $\omega = 10^9$ cycle/sec and $\omega = 2 \times 10^9$ cycle/sec. Hence if we neglect interference between the effect of scattering by the electrons and an intrinsic dispersion, the intrinsic dispersion in this frequency range is $\lesssim 10^{-9}$.

One source of a variation of speed with frequency would be if the photon rest mass were some nonzero number m_γ. In this case we would have

$$\frac{v}{c} = \sqrt{1 - \frac{m_\gamma^2 c^4}{\hbar^2\,\omega^2}} \simeq 1 - \tfrac{1}{2}\,\frac{m_\gamma^2\,c^4}{\hbar^2\,\omega^2} \quad (4)$$

(where $\hbar$ is Planck's constant) since the photon mass is known to be small compared to $\hbar\omega/c^2 \approx 10^{-40}$ g for $\omega = 10^8$ cycle/sec.

Thus the variation with frequency due to a photon mass is evidently greatest at small frequencies; as a result radio propagation is a much more sensitive test of this variation than optical propagation. The dependence on frequency is the same as that obtained from the plasma dispersion. Comparing the two, we get

$$\frac{v}{c} = 1 - \tfrac{1}{2}\left(\frac{m_\gamma^2 c^4}{\hbar^2} + \omega_p^2\right)\frac{1}{\omega^2} \quad (5)$$

The "index of refraction" is much greater for radio frequencies than for optical frequencies. We can therefore place an upper limit on the photon mass by assuming that a substantial part of the observed dispersion is due to photon mass, and by equating the photon mass and the plasma frequency with which the radio dispersion has been fitted

$$m_\gamma \leqslant (\hbar\,\omega_p)/c^2 = \hbar/c^2\sqrt{\bar{n}_e} \times 56 \times 10^3 = 6 \times 10^{-44}\sqrt{\bar{n}_e} \quad (6)$$

For the Crab Nebula pulsar, it is known (*4*) that $\bar{n}_e \lesssim 2.8 \times 10^{-2}$. Hence

$$m_\gamma < 10^{-44}\ \text{g} \quad (7)$$

If the interstellar electron density were known reliably, we could subtract this effect from Eq. 5 and obtain a better limit for m_γ. A still lower limit on the mass of the photon has been established by a study of the earth's magnetic field (*5*). This limit corresponds to

$$m_\gamma < 4 \times 10^{-48}\ \text{g}$$

However, the photons involved in the analysis of the earth's magnetic field are virtual photons with $\omega = 0$, rather than the real photons in the pulsar emissions, so that an independent measurement is still of interest. It would be useful in this connection if an extragalactic pulsar could be detected, since there is reason to expect that the electron density $\bar{n}_e$ is much lower between the galaxies; in this case an appreciably lower limit for m_γ could be established by measurement of the dispersion of an extragalactic pulsar.

No intrinsic variation of velocity with frequency other than that given in Eq. 4 is consistent with the theory of special relativity. However, since this theory should be checked whenever possible, it is of interest to ask what the data say about other variations. In general, we have

$$t = \frac{l}{v} = l\,\frac{dk}{d\omega} \quad (8)$$

where $k = k(\omega)$ is the wave number of a light wave of frequency ω.

It has been shown above that the effect of both interstellar plasma dispersion, or of a photon mass, can be represented by a term $+\,\omega_0^2/2\omega^2 c$ in $dk/d\omega$ when $\omega_0^2 \ll \omega^2$. Let us therefore write

$$\frac{dk}{d\omega} = \frac{\omega_0^2}{2\,\omega^2\,c} + \frac{\delta\,v\,(\omega)}{c^2} + \frac{1}{c} \quad (9)$$

where $\delta v(\omega)$ may be a noncovariant intrinsic dispersion. If we now compare the arrival times of an optical and a radio pulse, we obtain

$$\frac{t_R - t_{op}}{l} = \frac{\omega_0^2}{2\omega_R^2 c} - \frac{\omega_0^2}{2\omega_{op}^2 c} + \frac{\delta v_R}{c^2} - \frac{\delta v_{op}}{c^2} \quad (10)$$

We can safely neglect ω_0^2/ω_{op}^2, since ω_0^2 is approximately 10^9 from the above, and ω_{op}^2 is approximately 10^{30}.

It has been shown by Tanenbaum *et al.* (*6*) that the variation of arrival time within the radio region is well fitted by a ω_0^2/ω^2 law. This implies that the term $\delta v_R(\omega)$ is either zero or constant in that region, and also that ω_0^2 can be obtained from measurements on the radio pulses alone, independent of the origin of the dispersion.

Conklin *et al.* (*7*) have compared the arrival times of radio and optical pulses and have found that, after removal of the effect of interstellar dispersion, these arrival times agree to within 10^{-3} second. In our language, this means that

$$\left|\frac{\delta v_R - \delta v_{op}}{c^2}\right| = \frac{t_R - t_{op}}{l} - \frac{\omega_0^2}{2\omega_R^2 c} < \frac{10^{-3}\ \text{sec}}{l} \quad (11)$$

Hence

$$\left|\frac{\delta v_R - \delta v_{op}}{c}\right| < 6 \times 10^{-15} \quad (12)$$

which implies that any noncovariant part of the velocity of optical or radio electromagnetic radiation is less than one part in 10^{14} of the average velocity.

It would be useful to extend the comparison to higher frequencies by a consideration of the arrival times of optical and x-ray pulses.

G. FEINBERG

Department of Physics, Columbia University, New York 10027

References and Notes

1. See J. H. Sanders, *The Velocity of Light* (Pergamon, Oxford, 1965).
2. W. J. Cocke, M. J. Disney, D. J. Taylor, *Nature* **221**, 525 (1969).
3. B. Warner and R. E. Nather, *ibid.* **222**, 157 (1969).
4. D. H. Staelin and G. C. Reifenstein III, *Science* **162**, 1481 (1968).
5. A. S. Goldhaber and M. M. Nieto, *Phys. Rev. Lett.* **21**, 567 (1969).
6. B. S. Tanenbaum, G. A. Zeissig, F. D. Drake, *Science* **160**, 760 (1968).
7. E. K. Conklin, H. T. Howard, J. S. Miller, E. J. Wampler, *Nature* **222**, 552 (1969).
8. I thank Drs. R. Novick and A. Goldhaber for helpful conversations. Supported in part by the U.S. Atomic Energy Commission.

12 May 1969; revised 7 July 1969

Cynodont Reptile with Incipient Mammalian Jaw Articulation

Abstract. *A diagnostic mammalian character is jaw articulation between squamosal and dentary bones, replacing the quadrate-articular joint of reptiles. A newly discovered Argentinian Middle Triassic form shows, for the first time in an ancestral reptile, definite evidence of a squamosal-dentary articulation supplementary to the persistent primitive connection.*

Diagnostic characters separating reptiles and mammals have mainly to do with functional features or "soft" anatomy. There is, however, one osteological character highly useful to the student of phylogeny and paleontology. Mammals have abandoned the primitive jaw joint between quadrate and articular in favor of a new articulation between the squamosal bone of the skull and the dentary, which makes up the entire mammalian lower jaw. During this shift the half dozen other bones originally present in reptiles dwindled and disappeared from the jaw, and the quadrate and articular were transformed, together with the pre-existent stapes, into the mammalian series of ear ossicles.

Until recent years the reptile-mammal distinction in jaw articulation was clear-cut in the fossil record. The older, Mesozoic, fossil mammals are in general poorly known, but lower jaws are not infrequently found; in every case there is a distinct condyle on the dentary, obviously for articulation with the squamosal. Among mammal-like reptiles the dentary is much enlarged, but in every case reported the old quadrate-articular joint persisted.

In recent years the situation has become somewhat clouded. There is evidence that in at least some early mammals jaw elements other than the dentary were still present and that a vestigial quadrate-articular connection, in addition to a fully developed squamosal-dentary articulation, persisted for a time in some forms (*1*). Crompton (*2*) has described from the late Triassic Cave sandstones (? Rhaetic) of South Africa a form (*Diarthrognathus*) in which, in addition to the quadrate-articular joint, the dentary was in contact with the squamosal. This type, hence, is a structural intermediate, but it cannot be considered as an evolutionary reptile-mammal transition form, because true mammals were already in existence at the time of its appearance. The phylogenetic position of *Diarthrognathus* (whether reptile or mammal) is uncertain, and other features of its anatomy show that it is not affiliated with the typical cynodonts from which most, if not all, mammals descended.

Until the present the therapsid side of the picture has remained clear-cut. In the Lower Triassic of South Africa there are a number of carnivorous cynodonts (of which *Cynognathus* is best known) which quite surely represent an early stage in the reptile-mammal transition, in such features as the beginning of a secondary palate and enlargement of the dentary bone of the jaw with relative reduction of the other jaw bones. The articulation of the jaw, however, remains solely between quadrate and articular.

I am currently studying members of the family Chiniquodontidae, later representatives of this carnivorous cynodont line, present in the Middle Triassic of South America. Two genera of this family, *Chiniquodon* and *Belesodon*, from the Santa Maria beds of Brazil were described some time ago by von Huene (*3*). His material was so inadequate that little could be told of their structure; I am currently redescribing their skull structure from better material obtained by a Harvard expedition (*4*), and am, further, describing two additional genera from the slightly earlier Chañares beds of Argentina (*5*). The chiniquodontids show an advance over the Lower Triassic members of the group in certain regards, notably an increase in extent of the secondary palate to essentially the mammalian stage.

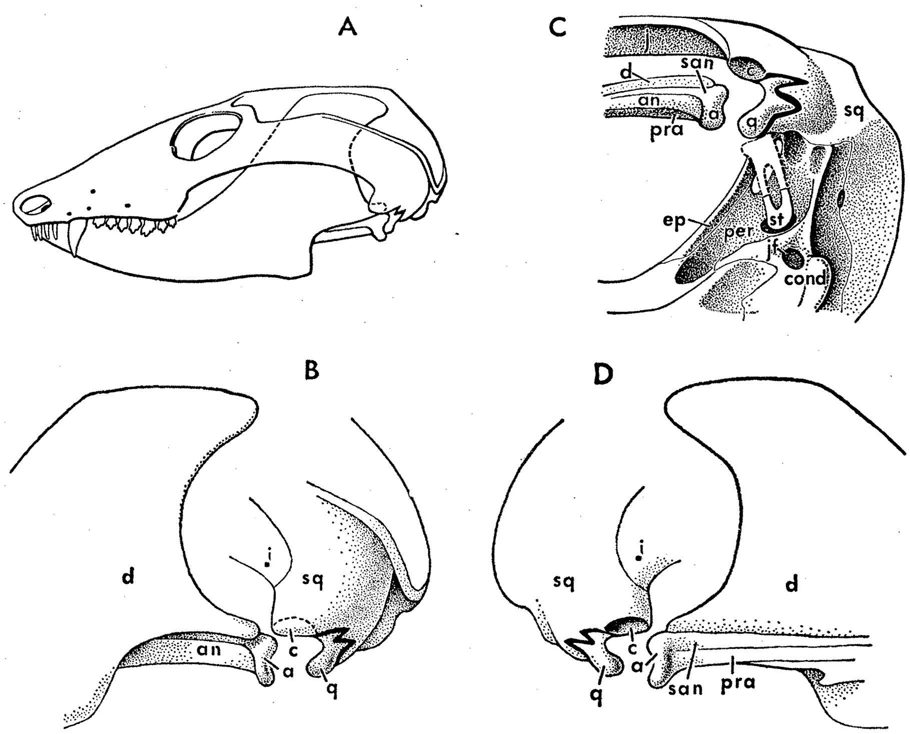

Fig. 1. (A) Side view of the skull of *Probainognathus*. (B), (C), and (D) Lateral, ventral, and medial aspects of the articular region of the left side of the skull and jaws. In each case the jaw is disarticulated and drawn slightly forward. Abbreviations are: *a*, articular; *an*, angular; *c*, incipient condyle on squamosal; *cond*, occipital condyle; *d*, dentary; *ep*, epipterygoid; *j*, jugal; *jf*, jugular foramen; *per*, periotic; *pra*, prearticular; *q*, quadrate; *san*, surangular; *sq*, squamosal; and *st*, stapes.