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Biological Modulation of the Earth's Atmosphere

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We review the evidence that the Earth's atmosphere is regulated by life on the surface so that the probability of growth of the entire biosphere is maximized. Acidity, gas composition including oxygen level, and ambient temperature are enormously important determinants for the distribution of life. We recognize that the earth's atmosphere deviates greatly from that of the other terrestrial planets in particular with respect to acidity, composition, redox potential and temperature history as predicted from solar luminosity. These deviations from predicted steady state conditions have apparently persisted over millions of years. We explore the concept that these anomalies are evidence for a complex planet-wide homeostasis that is the product of natural selection. Possible homeostatic mechanisms that may be further investigated by both theoretical and experimental methods are suggested.

> "The atmosphere, therefore, is the mysterious link that connects the animal with the vegetable, the vegetable with the animal kingdom." (Dumas and Boussingault, 1844).[1]

I. Introduction to Gaia: Le Milieu Exterior

The purpose of this paper is to develop the concept that the earth's atmosphere is actively maintained and regulated by life on the surface, that is by the biosphere. The ancient Greeks believed that all creatures on the earth, animals and plants, including man belonged to a common society. Their reference to this great communal being was "Gaia," or roughly, Mother Earth. We recognize that the earth's atmosphere differs greatly from that of the other terrestrial planets with respect to acidity, composition, redox potential, and temperature history as predicted from solar luminosity. This anomalous atmosphere has persisted over geological time periods. We believe that these properties of the terrestrial atmosphere are evidence for homeostasis on a planetary scale. In deference to the ancient Greek tradition, we refer to the controlled atmosphere-biosphere as "Gaia". We review some recent information about the terrestrial atmosphere in the light of the "Gaia hypothesis" (Lovelock, 1972; Lovelock and Margulis, 1973). First we consider the evidence that certain features of the terrestrial atmosphere (such as composition, temperature and pH) are under feedback control. We follow with suggestions for possible mechanisms involved in this complex homeostasis and ways of testing for them. We have written this paper to be comprehensible to a wide scientific audience, recognizing that an understanding of the earth's atmosphere will come only from cooperation of many scientists: planetary astronomers, geolo-

[1] See Aulie, 1970.

gists, meteorologists, chemists, physicists and biologists. Our purpose is to present the Gaia hypothesis for the reader's criticism.

II. The Atmosphere as Circulatory System of the Biosphere

A. The Earth and Its History had Life not Evolved

There are two ways of considering the abiotic atmosphere: one is to "delete" life from the present earth, the other to "interpolate" the Earth between Venus and Mars and consider an atmosphere on such an interpolated planet had life not evolved. If life vanished from the earth today, as chemists have calculated, the atmosphere would change. Nitrogen and oxygen, over oceans of water, are not an indefinitely stable mixture in the presence of thunderstorms and high energy radiation from the sun. In time, nearly all the oxygen and then the nitrogen in the air would disappear, leaving the element nitrogen in its chemically stable form: the nitrate ion in the seas. As long as free O would be provided by photolysis of water, nitrogen would be removed to the surface. The earth would become a planet more like Mars or Venus with an acid surface, an atmosphere mainly of carbon dioxide, but with traces of carbon monoxide, rare gases, and oxygen (Table I). As the reaction between nitrogen and oxygen and water vapor proceeded, the atmospheric nitrogen would be oxidized to nitric acid which would fall out as acid rain. Oxidative weathering would then remove oxygen down to very low levels (Walker, 1973b).

It is reasonable to assume that the atmospheric history of the earth, had life not evolved, would not have been greatly different from that of a planet interpolated between Mars and Venus in the present solar system. All three planets may have begun with reducing atmospheres, rich in hydrogen, methane and ammonia. Since the exospheric temperatures and the gravitational fields of these planets do not allow atomic hydrogen to remain, hydrogen was then lost to space. Some hydrogen chemically bound as NH_3, H_2O, CH_4, etc. would have persisted. Parallel with the loss of H_2 would be a change in the oxidation–reduction state of the atmosphere. Correspondingly, the planetary atmosphere and surfaces would go from very reducing to only slightly reducing and from ammonia-rich and alkaline to carbonate–nitrate–sulfate rich and acid. A model for the history of the gases of the atmosphere on an earth-like planet had life not evolved is shown in Fig. 1a. In contrast, Fig. 1b shows the historical course the atmosphere of the earth is thought to have taken with life present.

No planet, of course, on which the sun shines can ever be in thermodynamic equilibrium. The equilibrium of a lifeless

TABLE I

Atmospheres of the Terrestrial Planets (Pressures in Millibars)

Gas	Venus	Earth: Present	Earth: Model abiological[a] I	Earth: Model abiological[a] D	Mars
CO_2	90 000	0.3	0.3	1000	5
N_2	1000	780	< 1	30	< 0.05
O_2	0	210	≈ 1	0.3	< 0.1
Approximate total pressure in atmospheres	91	1	0.3	1	0.006

[a] Model dependent values: I, if the earth were interpolated between Mars and Venus and life had not evolved, D, if terrestrial life were deleted now. See text.

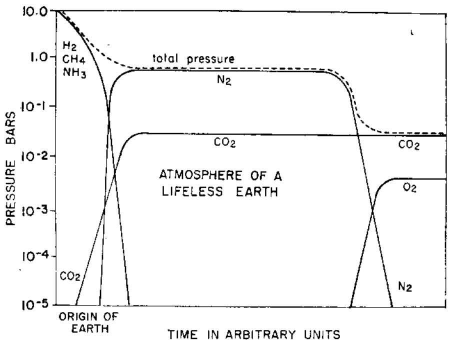

FIG. 1a. History of the gases of the atmosphere on a lifeless earth.

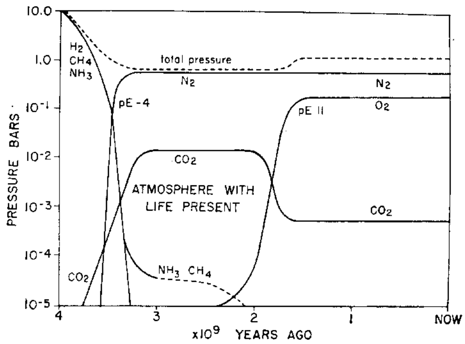

FIG. 1b. History of the gases of the atmosphere with life present.

planet which slowly changes as hydrogen is lost is better called abiological steady state. Thus, planets nearer the sun tend toward an atmosphere predominantly of carbon dioxide gas with some carbon monoxide and traces of oxygen and ozone. These are produced when sunlight dissociates carbon dioxide to CO and O and water to H and O; although the details of some processes are not entirely clear, H_2, O_2 and O_3 are formed by recombination. Since photodissociation occurs only at the outer reaches the bulk of the atmosphere is just carbon dioxide. If one assumes that similar initial conditions prevailed during the origin of Venus, Earth and Mars (with C/H ratios characteristic of chondritic meteorites), Venus, over its history, would have lost an ocean's worth of water and is further advanced to a final abiological equilibrium than the Earth. On this model liquid water has been lost from the surface and none is left to sustain the production of oxygen. As a consequence of the return to relatively more reducing conditions, high temperature and acidity which make the nitrate ion unstable on the Venus surface, nitrate there may have again reverted to N_2. It is possible that the photoproduced oxygen of Venus has gone to oxidize the surface rocks which may well have dissolved in the superheated steam. At temperatures greater than 350°C and pressures above 50 atmospheres, superheated steam dissolves aluminosilicate rocks. If this has been the case, Venus may turn out to have been melted to considerable depths and may harbor crystalline oxidized gem minerals in quantity. As the sun remorselessly continues to increase its output of energy, loss of hydrogen, utilization of surface oxygen for the oxidation of rocks, and thus a trend toward hot acid conditions would be expected to follow on a lifeless Earth and later on Mars. At no stage of the evolution of an abiotic planet would more than mere traces of atmospheric oxygen be expected and certainly with alkaline oceans present N_2 and O_2 would not coexist.

From astronomy, meteorology, physics, and equilibrium chemsitry, it is doubtful that we could have predicted the present environmental conditions on the earth. Given only these sciences, something is lacking in the understanding of the atmospheric history of our anomalous planet. The most conspicuous difference on the earth relative to the other terrestrial planets is the ubiquitous scum of the planet (and mostly lying within a few meters of the air, rock, water interface), namely the biota. Presumably it is this living system that is responsible for the phenomenon we are calling Gaia.

B. *What is Gaia?*

We believe that Gaia is a complex entity involving the earth's atmosphere, biosphere, oceans and soil. The totality constitutes a feedback or cybernetic system which seeks an optimal physical and chemical environment for the biota (Lovelock, 1972). The maintenance of relatively constant conditions by active control is conveniently described by the word "homeostasis," which we shall use henceforward for this purpose.

Man-made cybernetic systems require sensors, for example, the thermostat which senses room temperature. The sensor communicates instructions to a furnace or an air-conditioner so that if the temperature of the room changes from the point at which the thermostat is set, the system will respond by providing either cold or hot air so that the temperature stays constant. Cybernetic systems vary greatly in complexity but all require a number of essential elements: sensors, information storage, amplifiers, and feedback mechanisms. In our room thermostat example there first must be a way of telling what the temperature is and whether it has departed from the desired value (that particular value is information stored in the system). The output of energy from the sensor (e.g., the thermostat) itself is negligible—it could not possibly heat or cool the room. There then must be an energy amplifier, which in this example is the furnace that supplies the heat to the room or the air conditioner which removes heat. The heat that is supplied to or removed from the room causes a temperature change which feeds back to the thermostat, and when the thermostat receives a signal that the set point has been reached, an instruction is released so that the system turns off. This provides us with a closed loop and an example of what is meant by a cybernetic control system.

With biological cybernetic systems [see, for example, Riggs (1970) on the temperature control mechanisms of humans] there is not one but many methods of control used, e.g., sweating, shivering, the redistribution of blood flow, and heat production itself (Myers, 1971). The ultimate achievement of the desired core temperatures of a human in the face of environmental variation is more a matter of consensus among these control systems, acting via a particular brain Na^+/Ca^{2+} ratio (Myers and Tytell, 1972) than simple reference to a preprogrammed set point, as in our room thermostat example. We suspect that the earth's control systems follow a similar complex pattern more comparable to the temperature control in individual organisms than to man-made models. We want now to examine the consequences of our hypothesis that the earth's atmosphere–biosphere contains many cybernetic control systems responsible for maintenance of levels of temperature, acidity and composition tolerable to the earth's biota.

C. *A Long History of Constancy*

From the deposition of the earliest sediments, until the present it seems that the pattern of crustal alteration and sediment formation on the surface of the earth has not changed very much (Garrels and Mackenzie, 1971; Siever, 1973). The fossil record suggests the existence and lifetimes of ocean basins, lakes, and streams have not altered in three billion years. Rocks have been weathered and sediment deposited, soils have been made, transported, and lithified implying a consistent, dynamic relationship between forces set by the earth's physical conditions and mediated by the omnipresence of water and life. There is no evidence, at least in the last half billion years, that the oceans have altered much with respect to alkalinity, salinity or redox potential.

Temperature changes have occurred as the record of past glaciations attests. However, even at the farthest glacial advance, the mean temperature of the tropical regions was probably no more than 8°C lower than during interglacial periods (Emiliani, 1972). In general, ice ages affect most severely the temperature of regions above latitudes 45°N and below 45°S which constitute only 30% of the planetary surface. If the presence of some glaciation even during interglacial periods is assumed, the proportion affected is even less. Had the earth been entirely glaciated for tens of thousands or millions of years in the past, this fact would have been indelibly recorded and, in fact, episodes of widespread glaciation are relatively rare. Apparently, the temperature of the earth has not dropped sufficiently in the last 3.3 billion yr to cause complete freezing of all the oceans. As noted by Sagan and Mullen (1972; see below, Section III. D.) this is

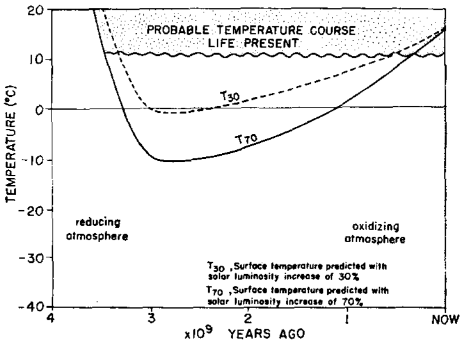

FIG. 2a. Temperature history of the earth: abiological prediction from solar luminosity.

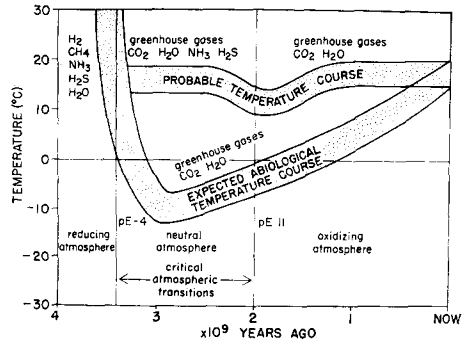

FIG. 2b. Temperature history of the earth: probable history derived from the fossil record.

remarkable when evaluated in the context of a fairly well established fact of astronomy: the energy output of the sun has been steadily increasing. Although the magnitude of increase is still a subject of debate, it has probably not been less than 40% and may have been as great as a threefold increase in the last 4 billion yr. Dilke and Gough, 1972, argue that the solar luminosity has increased 70%, and that the solar output fluctuates periodically over a range of 10% of its mean output, at intervals of less than 10^9 yr. But the surface temperature of the earth has not correspondingly risen and fluctuated as a function of solar luminosity, at least during the last billion yr. In fact, the surface temperature has probably not fluctuated more than 10°C around its present mean value. In Fig. 2 we compare the probable actual temperature history of the earth with that expected on an abiological model of the planet. The abiological model is based on two different estimates of the increase in solar luminosity: 30% (Sagan and Mullen, 1972) and 70% (Dilke and Gough, 1972: Fig. 2a and b). Manabe (1971), in a review of paleoclimatology, quotes a study suggesting the earth may be in an unstable climatic state. It can be calculated that a relatively small decrease in the solar output should cause ice cover to move southwards and more sunlight to be reflected to space, which should set up a positive feedback on cooling and possibly lead to permanent ice age conditions. Cooling might even proceed until the oceans froze. On the other hand Rasool and De Bergh (1970) have proposed that had the earth formed 6 million mi nearer the sun similar positive feedback processes would have led to the volatilization of the oceans, producing a runaway thermal condition leading to a high temperature and pressure atmosphere like that on Venus. That is, theoretically, approximately a 10% change in solar luminosity in either direction has been predicted to cause worldwide freezing or volatilization, something not observed, yet the data of astronomy imply a far greater change in solar luminosity than 10%, perhaps as great as 100%.

Not only has the sun's ouput been changing but the gas composition of the atmosphere and the nature of its surface has changed. These too have affected the amount of heat that the earth received from the sun. The probability that, by accident, the temperature has for 3.5 billion yr, without exception, followed the straight and narrow path optimal for surface life seems unbelievable. We conclude from the fact that the temperature and certain other environmental conditions on the earth have not altered very much from what is an optimum for life on the surface, that life must actively maintain these conditions.

D. Recognition of the Microbial Contribution

Until rather recently, the first 3000 million yr of earth history were thought to be without fossils or at least relatively lifeless. Traditionally the search had been for large fossils, but it is now realized that the ancient earth was teeming with prokaryotic microbial life. This understanding can be dated from the Tyler and Barghoorn report on the two-billion-year-old Gunflint chert (1954). Thin sections were made through precambrian sedimentary rocks; they revealed an extensive microbiota in which the organisms were composed of the primitive prokaryotic types of cells. Since then a great abundance of diverse fossil microorganisms has been recognized, some actually very well preserved (Schopf, 1972). As microbial ecologists become aware of the fossil record, there is a growing understanding of the microbial contribution to the formation of ancient as well as present day environments. The extent of the microbial contribution in the production and maintenance of the different gases of the contemporary atmosphere is shown in Table II. We assume their contribution to the ancient air was as significant as it is to the contemporary scene.

We emphasize the microbial contribution for two reasons: their metabolic versatility leading to profound environmental effects and because the regulation of the planetary environment was apparently proceeding long before the evolution of the larger (eukaryotic) life forms. Furthermore, nearly all the chemical transformations that large organisms perform microbes can also do. Eukaryotic plants and animals are metabolically very uniform. For example in prokaryotic

TABLE II

REACTIVE GASES OF THE EARTH'S ATMOSPHERE

Gas	Concentration ($\times 10^{-6}$)	Inorganic sources	Biological sources[a]	
			Gaian	Human
N_2	7.9×10^5	10^{-3}	1	0
O_2	2.1×10^5	1.6×10^{-4}	110	0
CO_2	320	10^{-2}	140	16
CO	0.08	$<10^{-3}$	1.5	0.15
CH_4	1.5	0	2	0
$(CH_2)_n$	0.001	0	0.2	0.2
N_2O	0.3	$<10^{-2}$	0.6	0
NH_3	0.006	0	1.5	0
NO_x	0.001	Total 0.7		0.16
H_2S	?	0	?	0
SO_2	2×10^{-4}	?	?	0.16
CH_3SCH_3	10^{-4}	0	0.2	0
CCl_4	10^{-4}	2×10^{-3}	0	0
CH_3I	10^{-6}	0	0.03	0
H_2	0.5×10^{-6}	1.6×10^{-4}	?	?

[a] $\times 10^9$ tons per year.

forms alone, including about 7500 different recognized species of blue-green algae, the typical plant and animal processes that interact with atmospheric CO_2 and O_2 (i.e., photosythesis and respiration) are well represented. The role of prokaryotic microorganisms in environmental alteration is well documented; they produce and break down the entire spectrum of naturally synthesized organic compounds and they release a wide range of gaseous end products such as methane, ammonia, volatile amines, organic acids, hydrogen sulfide, nitrogen, nitrous oxide, and so forth into the air (Alexander, 1961; Brock, 1966, 1970).

Fossil evidence for microbial activity is not limited to microbiota seen in thin section; a very important aspect is the interpretation of Precambrian stromatolites (Walter, 1972). These sediments are usually composed of calcium carbonate, although some are siliceous. They are often laminated structures (Fig. 3). Some are shaped like cones and some like cauliflowers. They are recognized now to be products of microbial activity. They represent sediment that was trapped and bound primarily by photosynthetic filamentous blue-green algae (Golubic, 1973). Evidence is accumulating that certain deep ocean stromatolites known as manganese nodules are the products of aerobic, nonphotosynthetic bacteria (Monty, 1974). They have a world-wide distribution, clearly forming the dominant biotic component of

TABLE II (*continued*)

REACTIVE GASES OF THE EARTH'S ATMOSPHERE

Residence time, years	Principal origins	Balance	References
10^6–10^7	Denitrifying bacteria	0	Robinson and Robins, 1971
10^3	Algal & green plant photosynthesis	0	Donahue, 1966
2–5	Respiration and combustion	+	Robinson and Robins, 1971
0.3	Methane oxidation, combustion minor; aerobic bacterial breakdown of heme	0	Wofsy *et al.*, 1972; Engel *et al.*, 1972
7	Anaerobic methane bacteria	0	Wofsy *et al.*, 1972
0.003	Green plants and industrial	?	Robinson and Robins, 1971
10	Denitrifying & ammonia oxidizing bacteria, heterotrophic bacteria and fungi	+	Robinson and Robins, 1971; Schutz *et al.*, 1970; Yoshida and Alexander, 1970
0.01	Bacteria and fungi	?	Robinson and Robins, 1971
0.01	?	?	Robinson and Robins, 1971
0.001	Anaerobic bacteria: *Desulfovibrio* and *Clostridia*	?	Grey and Jensen, 1972
0.01	Atmospheric oxidation, combustion	?	Robinson and Robins, 1971
0.003	Marine algae and green plants	?	Lovelock *et al.*, 1972
1	Atmospheric photochemistry	0	Lovelock, Maggs and Wade, 1973
0.003	Marine algae	0	Lovelock, Maggs and Wade, 1973
2	Photosynthetic bacteria, atmospheric methane oxidation, photolysis, not known	?	Brinkmann (see Ycas, 1972) Walker, J. (1973)

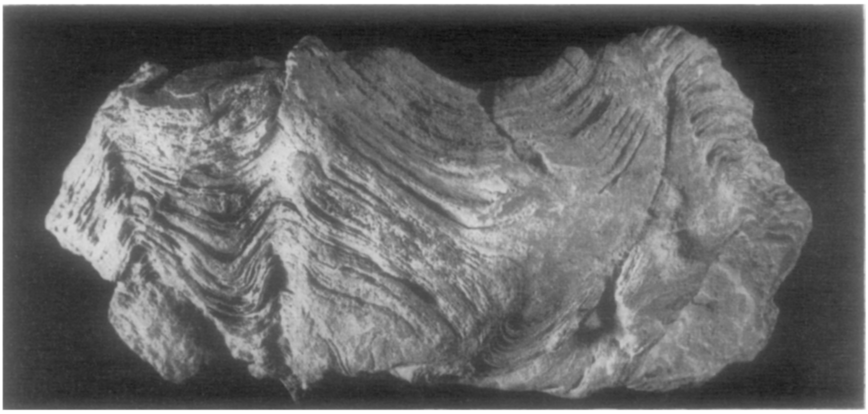

FIG. 3. Stromatolite: biosedimentary structure produced by $CaCO_3$ sediment trapping and binding activities of filamentous blue-green algae.

certain ancient ecosystems. From Fig. 4 (after Awramik, 1974) it can be seen that they flourished and reached their peak of diversity in the late Precambrian.[2] Some stromatolites have been detected in the Bulawayan sediments of South Africa, which may be as old as 3.3 billion yr (Kvenvolden, 1972; Barghoorn, 1973). The precise age of these earliest algal stromatolites may be questioned but certainly by the middle Precambrian (>two billion yr ago) stromatolites were well developed and widely distributed. The general environment of deposition in which 2-billion-year-old Canadian stromatolites were formed was similar to certain tropical environments today (e.g., Shark Bay, Australia), except for the conspicuous absence of metazoan animals and higher plants (P. Hoffman, cited in McAlester, 1968). Algal stromatolites are found in the most ancient fossil record, yet in the lower Precambrian, pebbles containing reduced minerals have been found suggesting they became weathered on the surface of an earth devoid of an oxygen containing

[2] Curve represents published descriptions of columnar stromatolites; twenty-five published reports are used. Radiometric ages of the boundaries are from Krylor (1972). The Pre-Riphean low point reflects the lack of work on stromatolites of that age. For example, it does not include stromatolites from the Gunflint Iron Formation and the Great Slave Lake (North America), the Transvaal Dolomite on the Venderdorp (South Africa). (See Aramwik, 1973 for details).

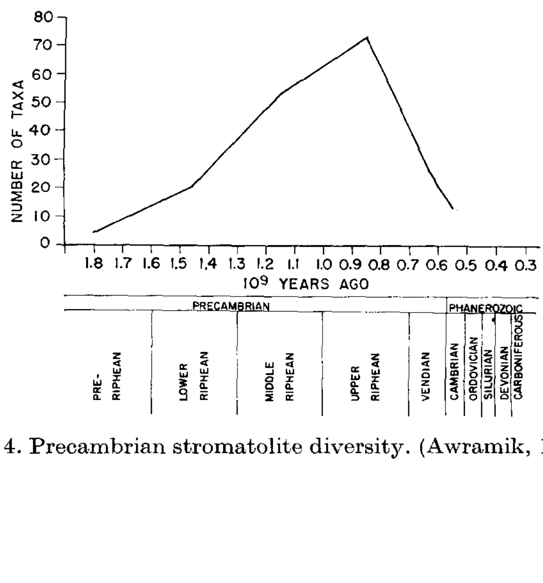

FIG. 4. Precambrian stromatolite diversity. (Awramik, 1973).

atmosphere. That is, blue-green algae were most likely excreting oxygen into the atmosphere at times when the atmosphere was composed of either the reduced gases or neutral ones such as N_2 and CO_2, The presence of Precambrian limestones and dolomites proves that CO_2 was in the atmosphere at pressures at least equal to those of today and probably higher. Even today many blue-green algae are found in anaerobic zones where, under very low oxygen tensions, they excrete oxygen. In the middle Precambrian iron minerals are laid down as banded iron formations (BIF's) that are thought to be influenced by microbial activity under relatively reducing conditions, whereas the iron minerals of the late Precambrian time to the present are primarily in "red beds", sedimentary deposits in which the iron is fully oxidized. It can be argued then that there have been two major transistions in the composition of the atmosphere: from highly reducing to neutral and from neutral to oxidizing. It has also been claimed that the formed earth never had a highly reducing atmosphere. [See Walker (1975) for discussion of these issues.] In full admission of these uncertainties, from astronomical deduction and interpretation of the fossil record, we have drawn an historical sequence of approximately three stages for the atmosphere (Fig. 2b): the formative earth and the lower Precambrian in which there is definite evidence for anaerobic conditions, the middle Precambrian in which there is evidence for the transition from reducing to oxidizing conditions (Cloud, 1972) and the late Precambrian until the present in which conditions have been oxidizing. According to Cloud, this last transition occurred between approximately 2.2 and 1.8×10^9 yr ago. It is highly likely that the transition to the oxidizing atmosphere was brought about by blue-green algal photosynthetic activity. At points of transition from reducing to oxidizing conditions there must have been profound changes in atmospheric composition. Reducing gases such as methane and hydrogen and possibly also ammonia must have been virtually eliminated, that is, reduced to their present level (Table II). The increase to the value of 20% oxygen may have involved some pressure change. We assume this transition placed a severe burden on the temperature regulating mechanisms of the planet, which we are hypothesizing. Yet in spite of this profound atmospheric change, average planetary temperature was maintained at that of liquid water and probably has not varied even tens of degrees.

We apply an analogous argument to the constancy of the alkalinity of the earth's surface. The suite of minerals laid down in acid lakes under acid conditions, for example in the volcanic lakes of Japan (Satake and Saijo, 1974; also see Brock, 1973) are quite different from those deposited under neutral and basic ones. The species of organisms associated with acid lakes also differ (Golubic, 1973 pers. comm). There may have always been local acid environments, as there are now, but in no geological period have they ever become the dominant environment of the earth. Clearly, in spite of drastic changes in solar luminosity and atmospheric composition there has been a maintenance of a neutral to alkaline temperate environment over long periods of time. Thus we believe that it is fruitful to assume that temperature, gas composition, and alkalinity have been actively modulated by organisms, especially microorganisms.

III. The Control Systems

A. What is Controlled?

Consider the problem of a planetary engineer whose contractor has given him a planet with a set of temperature and acidity specifications. This early planet of a cooler sun was covered by a reducing atmosphere and a life form that required a reducing environment, aqueous solution, neutral to alkaline pH's and temperatures between 0 and 50°C for its origin and early evolution. The engineer's job is to sustain these conditions for at least several billion years within limits acceptable to the surface life form. This implies the regulation of temperature, acidity and distribution of essential elements such as sulfur, phosphorus, and nitrogen. Let us consider for each in turn, the potential devices our engineer has at his disposal.

B. Temperature Control: Radiation Balance

Planetary surface temperatures may be altered by variations in the albedo and the emissivity of the planetary surface. Under present atmospheric conditions, a 1% change in albedo causes approximately a 1.7°C change in surface temperature according to the assumptions of Mathews *et al.* (1971). Changes of the absorption properties from the surface to the tops of the clouds may potentially alter albedo. Organisms such as grasses, trees, and algae actually alter absorbed and reflected light. With blue-green algae, for example, at least two types of induced changes in reflective properties of surfaces are possible; the first involves a physiological alteration of pigment production and the second an alteration in sediment trapping and binding. Blue-greens synthesize bright yellow pigments, carotenoids, and they also make deep reddish and bluish phycoerythrin and phycocyanin pigments. They always make both of these pigment classes which, together, may cause the reflection of a brown or even a black color. Variations in the amounts, proportions and exposure of these pigments influence the final surface qualities of algal mats. These algae can also sediment calcium carbonate which, of course, when pure, is chalky white. It is known that the surface properties of algal mats can change from light and reflective at one season to nearly totally black at another without any change in the composition of the algal community (Golubic and Awramik, 1973). Microbial communities of algae and bacteria that make up mats and stromatolites most likely respond to environmental variables that may change the reflective properties of shallow waters and land surfaces. This may also be true of green plant communities. Even the surface of snow may have its albedo altered by algal growth (Hardy and Curl, 1972),. Some very ancient limestones and siliceous cherts have enough elemental carbon to be almost black, even though their total quantity of carbon is far less than 1% (Barghoorn, private communication, 1973). Some microorganisms in the ancient past, and perhaps even now, may have set conditions where organic compounds oxidized to carbon black, which would serve as an ideal light absorber where warmth was needed. Clearly, trace quantities of minerals or carbon, for example, can make a vast difference in the color of the rock, and thus in heat balance. Albedo also can be strongly affected by cloud cover; see section F below.

C. Temperature Control: Emissivity

Changing the earth's albedo is not the only potential method available for control of surface temperature alteration. Another important surface property is emissivity. An object which is white or black in one spectral region is not necessarily white or black in another. Is it possible that the biosphere exercises some control of the emissivity? The effect on the radiative properties at a given wavelength is most significant when the interacting structures are of the order of a half or a quarter of a wavelength long. Thus in the 10 micron region structures in the range of 2.5 to 5μm could be quite important in altering emissivity properties in either direction, that is, toward greater or less absorption of the infrared radiation. Of course, 2.5 to 5μm is the typical size range for individual cells of blue-green algae and bacteria. Some of these organisms are as small as less than one micron and a little known fact is that blue-greens are sometimes as large as 60μm (Golubic, 1973, pers. comm.), but 2.5 to 5μm is the typical size at which these cells absorb visible light for photosynthesis. It has been recently realized that stromatolitic calcium carbonate deposits have a characteristic microstructure in the μm or tens of μm range. The microstructure is strongly influenced by the species composition of the cells that formed the stromatolite. This is only one small example. Ocean surface scum, forest canopy texture, soil particle size, and soil bacteria may affect the emissivity over large regions of the planetary surface, as well.

D. *Temperature Control: Gas Composition*

Another method of potential biological control of atmospheric temperature is via changes in gaseous composition of the atmosphere. Microorganisms of the soil are known to do much of the work in cycling the gases through the atmosphere; this can affect the surface temperature in a number of ways. The mere presence of a nonreactive gas in the atmosphere (which absorbs neither visible light nor infrared) will have little or no effect on the mean surface temperature. But the addition of gases composed of polyatomic molecules such as water vapour, carbon dioxide, or ammonia reduce the loss of radiation to space from earth by their infrared absorption. This is the well known "greenhouse effect" whereby the presence of carbon dioxide and water vapor in the current atmosphere keeps it tens of degrees above the temperature it would be were those gases not present.

The most effective greenhouse gases interact strongly with the biosphere. For example, not only is CO_2 the source of carbon for photosynthesis but it is removed from the atmosphere and converted into organic matter by many heterotrophic organisms [that is, nonphotosynthetic organisms that require organic compounds such as sugars or organic acids as food sources (Brock, 1970; Prevot and Fredette, 1966; Ljungdahl and Wood, 1969)]. Even animals incorporate atmospheric CO_2 and CO_2 is given off in respiration by nearly all organisms. Ammonia is another very active product of microbial metabolism. It is not only a product of fermentation but it is the breakdown product of purines, pyrimidines, and other organic compounds. The main nitrogenous excretory product of all organisms is ammonia, uric acid, or urea. Both uric acid and urea are easily broken down to ammonia so that the sources of ammonia are great in the biological world. The number of sinks is large also because nearly all bacteria and fungi utilize ammonia. Some bacteria oxidize ammonia (using gaseous O_2) to provide their major source of energy. These marine and soil bacteria convert ammonia to nitrite or nitrate which is then rapidly removed as a nutrient by photosynthesizers.

Thus many groups of organisms interact directly with the major greenhouse gases: ammonia, carbon dioxide, and water. The relative importance of ammonia with respect to CO_2 as a greenhouse gas which probably raised and maintained the temperature of the Precambrian above that of liquid water has been argued by Sagan and Mullen (1972). With respect to the third greenhouse gas, water, evaporation from the ocean, and the inorganic cycle in general is far more important than microbial metabolism.

E. *Temperature Control: Ozone*

Another gas, ozone (O_3), present in the atmosphere, may exert a subtle effect on temperature. Ozone is normally present only in very low quantities at the surface, but can be found at a concentration of 2 or 3ppm in the stratosphere.

Although there is no certainty that the mere presence of ozone in the stratosphere has any direct climatic significance, there is no doubt that the physical and chemical condition of the stratosphere is very different from that of an ozone-free stratosphere. If, for purposes of discussion, we assume a climatic significance for ozone, then we can further consider the role of gases such as nitrous oxide (N_2O) as ozone regulators. Nitrous oxide is a major biological product; hundreds of megatons a year are released by soil microorganisms.

It has long been known that "denitrifying" bacteria that convert soil nitrates and nitrites to N_2 produce N_2O as well, but it recently has been shown that N_2O production is far more widespread. Ammonia oxidizing bacteria such as *Nitrosomonas europaea* and ordinary heterotrophic bacteria and fungi (e.g., *Bacillus subtilis*, *E. coli*, *Aerobacter aerogenes*, *Aspergillus flavus* and *Penicillium atrovenetum*) release N_2O into the air. This release is inhibited by ammonia or high temperature (Yoshida and Alexander, 1970). The nitrous oxide so released decomposes in the stratosphere to give, among other products, nitric oxide, which is catalytically destructive of ozone. Alteration of the ozone layer may affect the height of the stratosphere and thus atmospheric circulation. This link between biological N_2O production and climate is admittedly tenuous. We introduce it primarily because in the context of Gaia the production of such large quantities of N_2O requires an expanation of its role in the atmosphere. It is very inert chemically except for its reactions in the stratosphere mentioned above. In addition, the analysis of gaseous nitrogen compounds eliminated by bacteria show that in the same culture of organisms the ratio of nitrogen to nitrous oxide can vary with environmental conditions. If nitrous oxide is a climatic regulator, then at least its production at the surface is responsive to climatic change.

F. Temperature Control: Particulate Matter in Suspension

There are three forms of particles in the atmosphere of potential importance in temperature control: clouds and mists of water droplets; particles, and liquid droplets produced by chemical reactions among gases; and particles of solid material, simply dust raised from the deserts. Let us first consider clouds. The area of the earth's surface at present is about 50% covered by clouds and because they are white, they reflect much sunlight back into space. If the proportion of clouds varied, it could affect the Earth's surface temperature quite significantly. If there were no clouds, it would be hotter and if there were a complete continual cloud cover, it would be a great deal colder. There is negative feedback on cloud cover, however. If the temperature rises there is more evaporation of water and more clouds form. If it falls there is less evaporation and fewer clouds form. There are several ways by which cloud cover via evaporation control could be altered by organisms. For example, the evaporation of water can be hindered by the accumulation of a monomolecular layer of steroids or other lipids. The application of such a monolayer of cholesterol has been found to be effective in reducing evaporation from reservoirs in summer. If a comparable process operated over the whole ocean, this might greatly affect cloudiness. Evapotranspiration by the trees of tropical forests pump huge quantities of water into the air; this strongly affects local cloud cover (Leith, 1971). Bacteria and algae tend to produce surface scums on ponds and slow running streams. This surface layer is composed of the cells and cell debris. Certain insects trap prey by secreting soap-like substances that alter the surface tension of water. Whether these sorts of examples are relevant on an oceanic scale is not obvious but clearly evaporation from puddles, ponds and lakes can be altered biologically and at least locally significantly alter humidity.

Particles can be formed when ammonia (which in our present atmosphere is entirely of biological origin) reacts with gases (which also may be biotic products). An ammonium sulfate aerosol in the stratosphere was discovered about 25 yr ago by Junge (1950). Such stratospheric aerosols apparently cause cooling which explains the drop in tropospheric temperature observed after major volcanic eruptions. Eruptions eject sulfur dioxide into the stratosphere which oxidizes to sulfuric acid and subsequently forms the ammonium sulfate aerosol. Unlike these natural stratospheric aerosols, tropospheric aerosols (those in the lower atmosphere, and more prevalent in the norther hemisphere) seem to be largely anthropogenic. The climatic significance of tropospheric aerosols (which seems to be determined by their optical properties such as color and refractive index) is still uncertain. If aerosols are colored, they absorb heat in the form of sunlight; such heat absorption offsets their back reflection of sunlight. It is not clear which process dominates. In any case the biotic (including anthropogenic) production of particulates is a potential factor in the regulation of temperature.

G. Control of Acidity

Acidity is another planetary property that, on the "Gaia hypothesis," requires cybernetic control. The earth seems to have maintained a pH remarkably close to 8 throughout history.

For example, it has been well established that blue-green algae, organisms responsible for stromatolite formation, do not tolerate conditions more acidic than about pH 4.5 (Brock, 1973). They do not grow at all below pH 4 and they do not generally grow well at pH's between 4 and neutral. During the Precambrian, they were widespread in the intertidal coastlines, worldwide, and probably spread to the subtidal and supertidal as well. The Precambrian distribution of stromatolites (see Fig. 4) and therefore the presence of blue-green algae argues that the marine continental shelves (at least from 3.3 to 0.5 billion yr ago) were neutral to alkaline then as they are today.

As the terrestrial planets become more oxidizing, and the reducing gases are removed by oxidation, their surfaces have become more acid. Yet, in spite of this tendency toward acidity with increasing oxidation of the atmosphere, the mean pH of the oceans has not dramatically fallen, or even changed much from its present value of 8.2. Attempts have been made to explain ocean buffering on the basis of both carbonate and silica equilibria (Sillen, 1966). We suggest that the ocean pH may be regulated instead by the biological production of ammonia. Approximately 2×10^9 tons/yr of ammonia are injected into the atmosphere. Unless the system is under cybernetical control, it is another remarkable coincidence that this is just a little more than sufficient to neutralize all of the acid formed in the oxidation of sulfur and nitrogen compounds (Table II).

There are a few regions in the world where acidity control is apparently not working properly. In Sweden and in northern United States the rain has been falling at about pH 3, and is said to be drastically reducing growth. Industrial SO_2 is presumably oxidized to sulfuric acid and is carried by the prevailing winds as a sulfuric acid aerosol to the affected regions. But the production of the neutralizing ammonia is temperature dependent and the acidity may be a consequence also of the recent temperature decline of northern regions. We interpret the unplanned experiment of releasing locally large quantities of sulfur oxides as a perturbation of the normal system of pH regulation. Measurements of the pH of the precipitation in Antarctica may help clarify the extent of the importance of the increase in industrial activity of western Europe in the production of acid rain.

H. Circulation of the Elements

The maintenance of the mass balance of the circulating elements may also be under active control. The circulation of essential elements amongst the growing species of organisms on the earth requires a fluid medium (whether an atmosphere or an ocean). It is doubtful if life could ever evolve on a planet with merely a solid surface and no atmosphere. Terrestrial vegetation can rapidly become depleted of necessary nutrient elements. The continual falling of pure water as rain on the surfaces tends to wash out these needed substances, yet they are apparently transferred back from the oceans to the land. The release of methylated derivatives of essential elements like iodine, sulfur, and perhaps selenium and phosphorus, we consider a mechanism of transporting these elements back to the land surfaces, a deliberate contrivance of Gaia to circulate them. Although the coastal areas would be the first recipients of limiting nutrients, due to the nutritional interconnections between terrestrial organisms (Atsatt, 1970), the rest of the land surfaces would eventually be "fed".

That many elements can be rendered volatile by methylation has been known for a long time, as people have found to their cost who have slept in bedrooms where the wall paper contained an arsenical pigment. Molds growing on the wall have released the arsenic as trimethyl arsine, sometimes fatally poisoning the sleepers (Alexander, 1961).

Inventories of the sulfur transfer through the atmosphere have been repeatedly made. Although uplifting and weathering resulting in cycling of sulfate in the form of evaporites occurs in about 10^8 yr, on the short term there has been a recognized deficiency of about 300 million tons (Kellogg *et al.*, 1972); more sulfur was being washed out by the rivers than could be accounted for from any source anywhere in the atmosphere or on the land. Challenger (1941) observed the emission of dimethyl sulfide by marine algae. The Gaia concept suggested we seek the quantities of dimethyl sulfide emitted by organisms, mainly algae, of the oceans of both the northern and southern hemispheres. When this was done we found the emissions were considerable and may account for this gap in the sulfur budget (Lovelock *et al.*, 1972). During the course of these sulfur measurements we also observed a large flux (30×10^6 tons/yr) of methyl iodide from the oceans (Lovelock *et al.*, 1973). Phosphorus is a vitally important element, unavailable biologically in its most common form: apatite. It is possible that trimethylphosphine or some other biologically released volatile is emitted from the ocean to restore phosphorus to the land. It may even turn out that airborne spores, birds, insects, and migratory fish are examples of "biologically released volatiles" of phosphorus. A present significant source of returned phosphorus is bird droppings onto the land (Hutchinson, 1950). This mechanisms of phosphorus circulation is obviously quite recent, that is primarily a Cenozoic phenomenon (0.07 billion yr old, whereas the need to circulate phosphorus is at least 2.7 billion yr old).

In the lower Precambrian it is likely that hydrogen sulfide, a direct product of microbial metabolism, was the gas that transported sulfur from the ocean to the land. The rate of oxidation of H_2S by dissolved O_2 at present in the open ocean is so rapid that there is no escape of the gas from the surface even though anaerobic muds at the sea bottom may be producing it in large quantities. In certain anaerobic ponds where the oxygen content of even the surface waters is low, H_2S may be emitted (Grey and Johnson, 1972), but this is rare relative to the widespread emissions of dimethylsulfide from the ocean surface. We assume the present dimethylsulfide cycle began during or after the transition to the oxidizing atmosphere (Lovelock *et al.*, 1972). The volatile substance dimethylsulfide may be further oxidized and distributed to the biota in the form of dimethylsulfoxide, under present oxidizing conditions.

I. Control of Redox Potential

Another problem of control, perhaps the least serious of all for life, is that of redox potential or pE. pE, analogous to pH, is a measure of electron concentration; it is defined as $pE = -\log [e^-]$. Thus a very low minus value of pE indicates an abundance of electrons and reducing material whereas a high positive value like a pE of $+13$ (the present value for the earth's surface) indicates highly oxidizing conditions. The earth's biota has adapted to a wide range of redox potentials. During its history the biosphere apparently has tolerated a transition from a pE of -8 to a pE of $+13$. Even though a drop in oxygen level is very serious from the point of view of people or any mammals, birds, or flowering plants, from a broad biological point of view free oxygen was one of the first pollutants. Even today oxygen is a poison to all organisms, including ourselves, at concentrations above those to which they are adapted (Haugaard, 1968). Many species are instantly put to death by parts per million of oxygen in water; these grow to large numbers when O_2 is locally removed. Life has apparently evolved adaptations and subsequently mechanisms to regulate this "pollutant" originally produced by photosynthetic life itself. Blue-green algae probably were the first organisms to eliminate oxygen as waste products of photosysthesis, and then adapt to its presence. Other anaerobic microbes responded by crawling out of the way of the oxygen into the muds. Later some, such as anaerobic pathogenic bacteria, responded by entering low oxygen tension animal tissues. All organisms have responded to the prevalence of free oxygen. Most animals and plants, including ourselves, eliminate CO_2 and H_2O utilizing O_2 to burn carbohydrates. (For the microbial evolutionary response to the elimination of oxygen by blue-green algae, see Margulis, 1972).

Thus the optimization of atmospheric oxygen at its present high levels must be a relatively recent product of evolution. It can be considered the outcome of greater fitness, in the Neodarwinian sense, of the aerobes in relation to the anaerobes. (For a discussion of the evolutionary mechanisms implied by the Gaia hypothesis, see Lovelock and Margulis, 1974). Today oxygen seems to be well-regulated and, in fact (because of the well-documented occurrence of large oxygen-requiring fossil organisms such as sharks), oxygen probably has not deviated drastically (more than a factor of two) from its present level since the late Precambrian. Several oxygen regulating mechanisms have been suggested, and in fact, many may be operative. The huge amount of methane production at the surface of the earth may ultimately favor the maintenance of high oxygen levels, as illustrated in Fig. 5 (Lovelock and Lodge, 1972). If O_2 levels locally become low enough the gowth of fermenting bacteria occurs, methane produced at the surface is transported through the tropopause of the stratosphere where it is oxidized to water which is photolyzed, H escapes and O_2 remains. As concerns the removal of oxygen, the large reserve of offshore continental shelf organic matter is likely to be a major oxygen sink (Walker, 1974b). Fire may be another "last resort" regulatory device, since the probability of spontaneous conflagration is a sensitive function of oxygen tension above organic matter (see Lovelock and Margulis, 1973, for discussion). In fact, regular forest fire may be a local mechanism to remove oxygen and nitrogen from the atmosphere to the biosphere in the form of nitrate. Apparently high nitrate concentrations found in soil following fire (at least in California chaparral) are due to the addition of ammonium and organic nitrogen in the ash, which become oxidized (Christensen, 1973).

We conclude that in the face of many potential perturbations, life has modulated the flow of energy and mass at the planetary surface. The temperature, *pE*, pH and element circulation seems, in general to be optimized for the whole of the interconnected biota. It seems to us unlikely

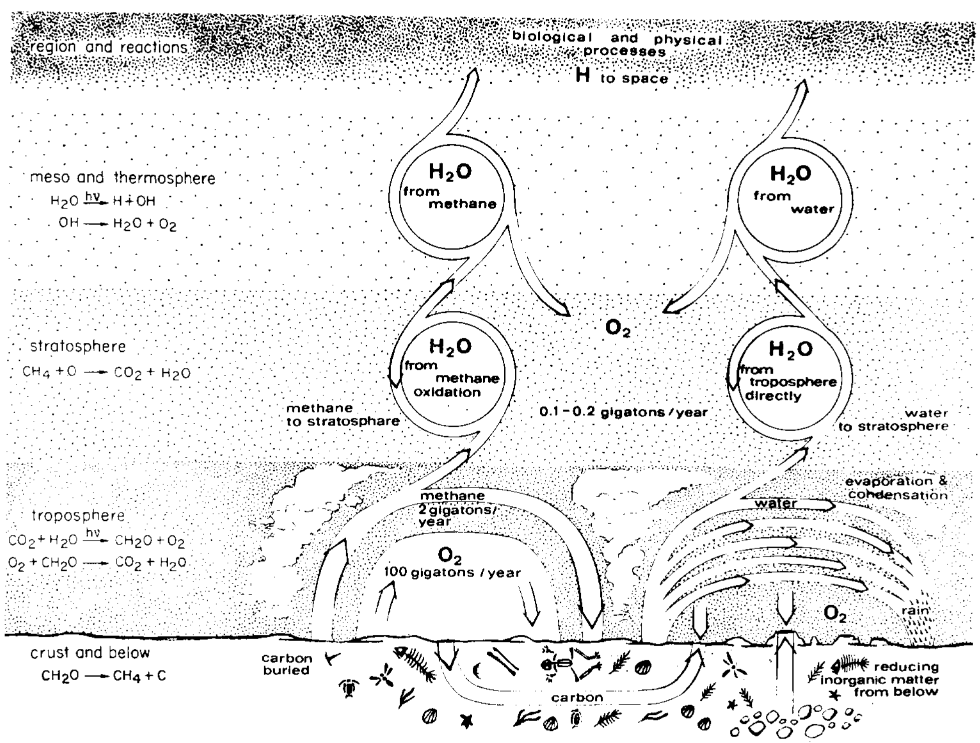

FIG. 5. Atmospheric oxygen maintenance from methane oxidation.

that these critical environmental variables are, by chance alone, precisely those required for the aerobic life that has evolved on earth. It is more reasonable to assume that, at the core at least (namely, the tropical and temperate zones) life has evolved and utilized many mechanisms to keep these variables from exceeding limits that are intolerable to all terrestrial species.

J. Early Evolution of the Control System

It is a recognized fact of biology that environments are regulated on a much more local scale, e.g., H_2 is emitted by purple nonsulfur photosynthetic bacteria to maintain a local reducing environment, termites build elaborate nests in which temperates and humidity are maintained at an optimum for the inhabitants; acids and bases are secreted by protozoa to regulate local pH's; fossil fuels are burned to maintain a semitropical environment in temperate zones by *H. sapiens.* We merely suggest the extrapolation of these ideas to the entire atmosphere-biosphere system. We are not claiming that a "planetary engineer" was actually commissioned but rather that Neodarwinian mechanisms of natural selection that have operated in the origin and evolution of the examples of local environmental control have also operated in the origin of these larger scale modulation mechanisms (see Lovelock and Margulis, 1974 for discussion). Life tends to grow until the supply of energy or raw materials set a limit. Probably a planet is either lifeless or it teems with life. We suspect that on a planetary scale sparse life is an unstable state implying recent birth or imminent death. (This predicts negative results for the Viking 1976 biological experiments. If no incorporation of $C^{14}O$ and $C^{14}O_2$ into organic compounds is detected under long Martian incubation conditions we would consider these significant negative results for the presence of life). Thus soon after life started on Earth probably the whole equatorial and temperate regions of the planet were colonized. The rapid and widespread alteration of the chemical environment consequent upon life's metabolism continually posed problems to the evolving species of the biosphere. As soon as metabolism induced physical conditions which were unfavorable, organisms were selected for which grew under such altered conditions. Subsequently new conditions provided opportunites for other variant forms. Those organisms able to maintain or alter conditions to favor their own growth have left more offspring. This evolutionary pattern imples that planetary homeostasis developed early in the history of the planet. Indeed the great crisis of the transition from an atmosphere in which such greenhouse gases as ammonia were stable to the more oxidized state where they were not, demands its establishment before 3×10^9yr ago (a time at and after which the planetary pE was greater than -4).

IV. Pleistocene Ice Ages: An Example

If our formulation is correct it becomes a problem to determine how changes in the environment can be "perceived" by the system Gaia and responded to. Which of the several possible mechanisms we have suggested, and many others we have not does the biota really employ to alter conditions on Earth? Can we prove the presence of a cybernetic system rather than a sequence of improbable events? It may take a long time to solve this multifaceted problem. However, in this context we offer for further study a reinterpretation of the Pleistocene ice ages as an example of partial failure of the temperature control system. Figure 6 shows the temperature

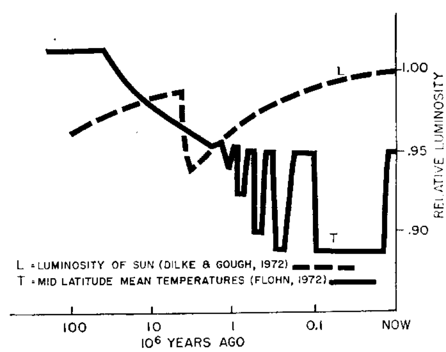

FIG. 6. Mid-latitude paleotemperatures compared with solar luminosity.

changes in the mid-latitudes of the earth over the last 100 million yr. Although the mid-latitude temperatures have fluctuated between about 0 and 20°C the temperature of the tropical regions probably hasn't changed by more than several degrees. As this diagram shows the temperature on the whole has been tending to fall in spite of the fact that the sun has been increasing its output.

If we erased temperature on the vertical and time on the horizontal axis our hypothetical planetary engineer would probably recognize this as a chart of the behaviour of an unstable control system in which instability had developed leading to oscillation yet control had not failed altogether (Fig. 6). These oscillations beginning just prior to a million years ago, of course, were the Pleistocene ice ages. By the Gaia hypothesis these temperatures oscillations might represent a partial failure of homeostatis which will be understood only when the basic thermostatic mechanism is known.

Apparently there were three major critical times at which extensive ice ages have been documented: the middle Precambrian, the Permocarboniferous boundary, and the recent series of Pleistocene glaciation episodes, as shown in Fig. 6. Although perhaps fortuitous, a possible correlation between evolutionary crises and ice ages deserves further study. The first one may have been related to the transition to an oxidizing atmosphere which must have posed an enormous problem to the whole terrestrial temperature control system. Permocarboniferous glaciation represented a time at which there was a great transition from aquatic to terrestrial environments. That is, that was a time at which (following the evolution in the Mississippian, 345 million yr ago, of the seed habit; Banks, 1970) the land became covered with macroscopic photosynthetic life forms on the scale of forests, and then of course, the evolution of terrestrial animals followed. The Cenozoic temperature decline may be related to replacement of conifers by decidious forests or perhaps the origin of the Hatch-Slack (C_4) pathway of CO_2 fixation in angiosperms (Welkie and Caldwell, 1970). These efficient plants, for example many grasses, are able to remove atmospheric CO_2 down to virtually zero (<2 ppm) relative to conventional Calvin dark cycle plants (C_3) that can only photosynthesize at minimal levels of CO_2 of about 70 ppm. Regardless of the mechanism regular periodicity of oscillation of this type is characteristic of an amplified system subject to positive feedback control.

The Gaia model requires many feedback loops and with different time constants; they are needed to cope with the maintenance of the temperature and composition of the atmosphere of the whole planet. The slow temperature changes due to solar luminosity increase need only a comparably slow set of compensating mechanisms. Other changes may require immediate short term responses. The lack of temperate control seen in Fig. 6 probably relates to only one system. If all systems had failed, the earth might either have frozen or approached boiling for it is poised on a razor's edge of temperature control (see Section II.C.).

Apart from its entertainment value, the Gaia hypothesis is principally useful in suggesting experimental questions in many scientific disciplines. The heuristic value implied, as well as the requirement for co-operation of differently trained experts, itself may justify the hypothesis. And possibly the Gaia hypothesis may eventually provide a true description of the atmosphere of our anomalous planet Earth.

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References

Alexander, M. (1961). "Introduction to Soil Microbiology," John Wiley & Sons, N.Y.

Atsatt, P. R. (1970). Hemiparasitic flowering plants: phenotypic canalization by hosts. *Nature* **225**, 1161–1163.

Aulie, R. P. (1970). Boussingault and the nitrogen cycle. *Proc. Amer. Phil. Soc.* **114**, 435–478. Quote on p. 452.

Awramik, S. M. (1973). Ph.D. Thesis. Harvard University, Department of Geology, Cambridge, Massachusetts.

Banks, H. P. (1970). Major geological events and the geological record of plants. *Biol. Rev.* **47**, 451–454.

Barghoorn, E. S. (1973). *In* "Origins of Life: Chemistry and Radioastronomy" (L. Margulis, ed.). Springer-Verlag, Heidelberg and New York.

Brock, T. D. (1966). "Principles of Microbial Ecology." Prentice-Hall, Englewood Cliffs, New Jersey.

Brock, T. D. (1970). "Biology of Microorganisms," Prentice-Hall, Englewood Cliffs, New Jersey.

Brock, T. D. (1973). Lower pH limit for the existence of blue-green algae: evolutionary and ecological implications. *Science* **179**, 480–483.

Challenger, F. (1951). Biological methylation. *In* "Advances in Enzymology," Vol. 12, p. 429–491.

Christensen, N. L. (1973). Fire and the nitrogen cycle in California chaparral. *Science* **181**, 66–68.

Cloud, P. E., Jr. (1968a). Atmospheric and hydrospheric evolution on the primitive earth. *Science* **160**, 729–736.

Cloud, P. E., Jr. (1968b). Premetazoa evolution and the origin of metazoa. *In* Evolution and Environment" (E. T. Drake, ed.), pp. 3–72. Yale Univ. Press, New Haven, Conn.

Cloud, P. E., Jr. and Semikhatov, M. A. (1969). Proterozoic stromatolite zonation. *Amer. J. Sci.* **267**, 1017–1061.

Dilke, F. W. W. and Gough, D. O. (1972). The solar spoon. *Nature* **240**, 262–263.

Donahue, T. M. (1966). The problem of atomic hydrogen. *Ann. Geophys.* **22**, 175–188.

Engel, R. R., Matsen, J. M., Chapman, S. S., and Schwartz, S. (1972). Carbon monoxide production from heme compounds by bacteria. *J. Bacteriol.* **112**, 1310–1315.

Emiliani, C. (1972). Quaternary paleotemperature and duration of the high temperature intervals. *Science* **178**, 398–400.

Garrells, R. and MacKenzie, F. (1971). "Evolution of Sedimentary Rocks." W. W. Norton, N.Y.

Giffen, C. E. and Lovelock, J. E. (1969). Planetary atmospheres: compositional and other changes associated with the presence of life. *Advanced Space Experiments* **25**, 179–193.

Grey, D. C. and Jensen, M. L. (1972). Bacteriogenic sulfur in air pollution. *Science* **177**, 1099–1100.

Golubic, S. (1973). The relationship between blue-green algae and carbonate deposits, *In* "Biology of the Blue-Green Algae," N. G. Carr and B. A. Whitton (eds.), Blackwell Scientific Publications, Ltd. Oxford. pp. 434–472.

Golubic, S. and Awramik, S. (1974). (Description of an algal mat on Cape Cod, in preparation).

Hardy, J. T. and Curl, H., Jr. 1972. The candy-colored, snow flaked alpine biome. *Natural History* **81**, 75–79.

Haugaard, N. (1968). Cellular mechanisms of oxygen toxicity. *Physiol. Reviews* **48**, 311–72.

Hutchinson, G. E. (1950). Survey of contemporary knowledge of biogeochemistry. 3. The biogeochemistry of vertebrate excretion. *Bull. Amer. Museum Nat. Hist.* **96**, 1–554.

Junge, C. E. (1950). Sulphur in the atmosphere. *J. Geophys. Res.* **65**, 227–237.

Kellogg, W. W., Cadle, R. D., Allen, E. R., Lazarus, A. L., and Martell, E. A. (1972). The sulfur cycle. *Science* **175**, 587–596.

Kushner, D. J. (1971). Life in extreme environments. *In* "Chemical Evolution and the Origin of Life" (R. Buvet and C. Ponnamperuma, eds.) p. 485–491. North Holland Publishing Co., Amsterdam and London.

Kvenolden, K. (1972). Organic geochemistry of early Precambrian sediments: 24th International Congress, Section 1, p. 31–41.

Leith, H. 1972. Modeling the primary productivity of the world. *Turk. Biol. Derg.* **22**, 85–88, and personal communication.

Lovelock, J. E. (1972). Gaia as seen through the atmosphere. *Atmos. Environ.* **6**, 579–580.

Lovelock, J. E. (1971). Air pollution and climatic change. *Atmos. Environ.* **5**, 403–411.

Lovelock, J. E. and Lodge, J. P. (1972). Oxygen in the contemporary atmosphere. *Atmos. Environ.* **6**, 575–578.

Lovelock, J. E., Maggs, R. J., and Rasmussen, R. A. (1972). Atmospheric dimethylsulfide and the natural sulfur cycle. *Nature* **237**, 452–453.

Lovelock, J. E. and Margulis, L. 1973. Atmospheric homeostasis by and for the biosphere: the gaia hypothesis. *Tellus*. In press.

Lovelock, J. E. and Margulis, L. (1974). Homeostatic tendencies of the earth's atmosphere. Proceedings of the first ISSOL meeting. *Origins of Life*. **1**, 12–22.

Lovelock, J. E., Maggs, R. J. and Wade, R. J.

(1973). Halocarbons in and over the Atlantic. *Nature* **241**, 194–196.

LJUNGDAHL, L. G. AND WOOD, H. G. (1969). Total synthesis of acetate from CO_2 by heterotrophic bacteria. *Ann. Rev. Microbiol.* **23**, 515–538.

MANABE, S. (1971). *In* "Man's Impact on the Climate" (W. H. Matthews, W. W. Kellogg and G. D. Robinson, eds.), pp. 249–264. M.I.T. Press, Cambridge, Mass.

MARGULIS, L. (1972). Early cell evolution. *In* "Exobiology" (C. Ponnamperuma, ed.) pp. 342–368. North Holland Publishing Co., Amsterdam and London.

MATTHEWS, W. H., KELLOG, W. W., AND G. D. ROBINSON, eds. (1971). "Man's Impact on the Climate." M.I.T. Press, Cambridge, Mass.

MCALESTER, A. L. (1968). "The History of Life." Prentice-Hall, New York.

MONTY, C. V. (1974). Manganese nodules are bacterial oceanic stromatolites. *Ann. Soc. Geol., Belgique.* In press.

MYERS, R. D. (1969). Temperature regulation: neurochemical systems in the hypothalamus. *In* "The Hypothalamus: Anatomical, Functional, and Clinical Aspects," pp. 506–522. Charles Thomas, Springfield, Illinois.

MYERS, R. D. (1971). *In* "Pyrogens and Fever" (G. E. W. Wolstenholme and J. Birch, eds.), p. 144. Churchill, London.

MYERS, R. D. AND TYTELL, M. (1972). Fever: reciprocal shift in brain sodium to calcium ratio as the set-point temperature rises. *Science* **178**, 765–769.

PREVOT, A. R. AND V. FREDETTE. (1966). "Manual for the Classification and the Determination of the Anaerobic Bacteria." Lea and Febiger, Philadelphia.

RASOOL, S. AND DEBERGH, C. (1970). The runaway greenhouse and the accumulation of CO_2 in the Venus atmosphere. *Nature* **226**, 1037–1039.

RIGGS, D. S. (1970). "Control Theory and Physiological Feedback Mechanisms." The Williams and Winkins Co., Baltimore.

ROBINSON, E. AND ROBINS, R. C. (1971). *In* "Global Effects of Environmental Pollution" S. F. Singer, ed.). Reidel, Dordrecht, and Springer-Verlag, New York.

SAGAN, C. AND MULLEN, G. (1972). Earth and Mars: evolution of atmosphere and surface temperatures. *Science* **177**, 52–56.

SATAKE, K. AND SAIJO, Y. (1974). Carbon dioxide content and metabolic activity in some acid lakes in Japan. *Limnol. Oceanogr.* **19**. In press.

SCHOPF, J. W. (1972). Precambrian paleobiology. *In* "Exobiology" (C. Ponnamperuma, ed.), pp. 6–61. North Holland Publishing Co., Amsterdam and London.

SCHUTZ, K., JUNGE, C., VECK, R. AND ALBRECHT, B. (1970). Studies in atmospheric N_2O. *J. Geophys. Res.* **75**, 2230–2246.

SIEVER, R. (1973). Oral comments *In* "Origins of Life: Chemistry and Radioastronomy: The Fourth ICP conference on the Origins of Life" (L. Margulis, ed.). Springer-Verlag. N.Y.

SILLEN, L. G. (1966). Regulation of O_2, N_2 and CO_2 in the atmosphere: thoughts of a laboratory chemist. *Tellus* **18**, 198–206.

TYLER, S. A. AND BARGHOORN, E. S. (1954). Occurrence of structurally preserved plants in Precambrian rocks of the Canadian shield. *Science* **119**, 606–608.

WALKER, J. C. G. (1973). Molecular hydrogen in the troposphere. (Manuscript).

WALKER, J. C. G. (1974). Stability of atmospheric oxygen. *Amer. J. Sci.* In press.

WALKER, J. C. G. (1975). Evolution of the atmosphere. (Manuscript in preparation.)

WALTER, M. R. (1972). Stromatolites and the biostratigraphy of the Australian precambrian and Cambrian. Special paper in *Paleontology* (11). The Paleontological Association, London.

WELKIE, G. W. AND CALDWELL, M. (1970). Leaf anatomy of species in some dicotyledon families as related to the C_3 and C_4 pathways of carbon fixation. *Canad. J. Botany* **48**, 2135–2146.

WOFSY, S. C., MCCONNELL, J. C., AND MCELROY, M. B. (1972). Atmospheric CH_4, CO and CO_2. *J. Geophys. Res.* **77**, 4477–4493.

YOSHIDA, T. AND ALEXANDER, M. (1970). Nitrous oxide formation by *Nitrosomonas europaea* and heterotrophic microorganisms. *Soil Science Soc. Amer. Proceedings* **34**, 880–882.

YCAS, M. (1972). Biological effects on the early atmosphere. *Nature* **238**, 163–164.