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Historical Review

The carbonaceous chondrites have occupied an interesting place in the study of meteorites for many years. Some 17 stones belonging to this general classification are known and authorities agree on their classification, but perhaps another half-dozen might be included in this type of stone. They contain carbon in easily detectable amounts, and in some of them there is easily recognized carbonaceous material. They represent thus some three per cent of the total observed falls, and though they are a minor fraction of the total they are far from being negligible in number.

The first stone of this class was observed to fall on March 15, 1806, in Alais, France. This stone was sent to Berzelius who in 1834 examined it chemically. He appears to have been the first scientist of reputation to have remarked about the similarity of its carbonaceous compounds to terrestrial biological material, and he asked the question, "Does this carbonaceous earthy material truly contain humus or a trace of other organic compounds? Does this possibly give a hint concerning the presence of organic structures in other planetary bodies?" This question has been asked repeatedly during the years since, but possibly only in very recent years have the techniques developed to a point where a definite answer can be given.

Wöhler and Hörnes in 1859 investigated another stone, Kaba, which fell in Hungary, and as a result of their work they believed that the

carbonaceous compounds were of biological origin. In 1864 a stone fell near the village of Orgueil in France. The stones were picked up immediately after the fall and were analyzed by Daubree and Cloëz. The fresh stone contained an ammonium salt, probably ammonium chloride, which was reported to accumulate on the fused crust of the stone in the course of time. Cloëz found organic matter comparable to peat, lignite, and organic matter in soils. The total carbonaceous material amounted to 6.4% according to these workers, and this is similar to that reported by Wiik in 1956, though the two types of analyses are not exactly comparable.

Berthelot examined the Orgueil meteorite in 1868 and reported material similar to petroleum of the general formula $C_n H_{2n+2}$. In fact, it was his conclusion that the compounds were hydrocarbons. It is interesting to note that during the nineteenth century outstanding chemists investigating the material in these meteorites reported material similar to that found on earth which is confidently ascribed to decomposition products of biological material.

Not until 1953 did Mueller again undertake to investigate the carbonaceous material in these objects. He investigated the Cold Bokkeveld meteorite and reported that carbonaceous material containing carbon, nitrogen, oxygen, hydrogen, sulfur and chlorine could be extracted from this stone. This latter observation is somewhat different from that of the earlier investigators for material of this composition would hardly correspond to hydrocarbons as reported earlier. Of course any extract is likely to consist of a mixture of chemical compounds.

During recent years careful studies have been made of the chemical composition of the inorganic materials. Berzelius observed clay-like minerals and this observation has been confirmed by later workers. Pisani, 1864, and Cloëz, 1864, independently demonstrated the presence of soluble

salts of ammonium, potassium, magnesium and sodium with sulfate and chlorine as the anions. Ammonia was not found by Wiik and it probably has escaped from the stone during the century of storage.

The Orgueil meteorite contains considerable amounts of water. Wiik reported 19.89% but earlier observers found somewhat less. It appears that these stones absorb water from the atmosphere in variable amounts, though Boato showed on the basis of the isotopic composition of the water extracted that that removed at higher temperatures is definitely different from any terrestrial sources of water. It appears therefore that some of the water has exchanged with terrestrial atmospheric water but that part of the water has remained in these stones from the time of fall. The difference in isotopic composition is not so great but what chemical ~~fractions of isotopes~~ ^{fractionation of the isotopes} of hydrogen may account entirely for the difference in the observed compositions.

In 1924 Spielmann suggested that the carbonaceous materials were produced by the action of terrestrial water on indigenous metal carbide minerals in the meteorites. But no one has ever found carbides in the carbonaceous chondrites and the observation that fatty acids are present in these meteorites as well as other complicated compounds to be discussed later shows definitely that this cannot be the origin of these compounds.

Mineral and Inorganic Chemical Composition

In more recent years Nagy and Anders and their coworkers have studied the mineral composition and have reported the presence of ferrous magnesium carbonate, calcium magnesium carbonate, and magnesium sulfate. They have concluded that at least certain of these meteorites consist of original mineral mixtures which have been acted upon by water.

The chemical composition of these objects varies considerably with respect particularly to the content of iron, water, sulfur, and carbon, and therefore in order to make a comparison with other objects in regard to the

more abundant elements, analyses calculated with the sulfur, carbon, and water eliminated are listed in Table 1. The columns give the chemical analyses of the Orgueil meteorite, the Mighei meteorite, and the average of the high iron group chondrites. The analyses of Orgueil and Mighei are from the work of Wiik who classified the carbonaceous chondrites into three groups - Type 1 containing 15-20% of water and no metal, Type 2 containing about 10% of water and very small amounts of metal, and Type 3 containing considerable amounts of metal, very little water, and small amounts of carbon. This last group has been reclassified by Mason as the olivine-pigeonite chondrites. Orgueil and Mighei are Types 1 and 2 respectively. It will be noted from Table 1 that the composition of the carbonaceous chondrites is very similar to that of the high iron group chondrites as classified by Urey and Craig with respect to other elements than sulfur, carbon, and water. It will be noted, however, that the ratio of iron to silicon is slightly higher in the chondritic meteorites of Type 1 than is true for the high iron group chondrites as a group. Thus the concentration of iron is exceptionally high in these objects.

Table 2 gives the percentage of sulfur, water, and carbonaceous material according to Wiik's analyses. We thus see that there are substantial amounts of sulfur and water and carbonaceous material in these objects.

The salts observed are those that are characteristic of what we expect the primitive oceans of the earth to have been. In the absence of an oxidizing atmosphere we would expect to find such reduced substances as the ferrous ion, ammonium ion, sulfide, and sulfur, though as time progressed we expect that carbonaceous material was oxidized to carbonate and sulfide to sulfate. As has been pointed out elsewhere, we can expect that hydrogen peroxide would be produced by ultraviolet light on such a primitive earth,

and Lewis has shown that hydrogen peroxide will produce both sulfur and sulfate from sulfide. We can expect that one of the first oxidation products of iron acted upon by water will be magnetic iron oxide which is also observed in these objects. The minerals observed are indeed those that would be characteristic of a primitive ocean, but the chemical composition shows that no sorting by running water has occurred since the composition is so similar to that of other meteorites which approximate to our ideas of the primitive abundances of the elements.

The mineral structure of the Orgueil meteorite is quite remarkable. In Figure 1 we see a mineral grain at the lower right-hand side, and a vertical magnesium sulfate vein toward the middle region of the picture. The other minerals are similar to clays. This microphotograph is 0.7 mm. wide. In Figure 2 we see some lath-like structures very similar to clay structures. These structures are very similar to what are called pyroclastic sediments - that is, sediments of volcanic ash or something of this sort settling in water. This indicates that this material has had a low temperature history and in physical appearance is similar to terrestrial ash flows, although the Orgueil meteorite has a completely different chemical composition, since terrestrial ash flows have an approximately granitic composition which is much higher in the constituents aluminum oxide, silicon oxide, and calcium oxide, and is lower in magnesium than is true of this meteorite. These mineralogical studies have been made by DuFresne and Anders and by Nagy.

Reports of Biological Material

In 1961, Nagy, Meinschein, and Hennessy presented results of their studies on carbonaceous chondrites to the New York Academy of Sciences, and this paper and their subsequent work raised much controversy in regard to the character of the organic material present in these objects. They had been working on the organic chemistry of petroleum and found on the basis of tests similar to

those which they had been using in this field that the material in the Orgueil meteorite was very similar to terrestrial fossil biological material. Students of meteorites including the present writer were immediately certain that this could not be true, because of the mineral chemical composition discussed above which strongly indicates that no sorting of material as found in sedimentary rocks had occurred. It is difficult to believe that biological material could have evolved in the absence of water and hence the inorganic constituents should have been subjected to the sorting effects observed as a result of the presence of water. This reaction of specialists in the field of meteorites came as a great surprise to Nagy, Meinschein, and Hennessy. They thought that they had made an interesting discovery and that there would be little opposition to its acceptance.

The first analysis made by these authors involved the extraction of organic material from the Orgueil meteorite and the subjection of this material to mass spectrographic analysis as is regularly used in studying biological material in soils, petroleum, and materials of this kind. They believed that they found the same pattern of molecular fragments in the meteorite as they observed in terrestrial fossil biological material.

Subsequent to this work Claus and Nagy observed small rounded objects in this meteorite which they interpreted as fossil microorganisms which they classified into various groups. Some were of quite simple structure while others were of a very complicated kind. Staplin who is a micropaleontologist working on problems of petroleum deposits also found objects which he interpreted as fossils. There is some similarity between the two groups of objects observed by these men. Anders and Fitch showed quite conclusively that some of the observed forms were indeed recent contaminants, namely, pollen grains. They found that some of the more complicated structures could be produced by

the staining techniques, which had been used, acting upon the ragweed pollen. However, it seems certain today that the more simple forms that have been observed are indeed indigenous to the meteorite, but of course such simple forms may indeed be artifacts rather than fossils.

Working with the Mighei meteorite, which is a Type 2 carbonaceous chondrite, Timofejev also extracted what he believed to be fossilized objects of a type similar to an alga type of object such as the dinoflagellates.

Figures 3, 4, 5, 6 and 7 show reproductions of various objects that have been reported in the Orgueil and Mighei meteorites. Many other examples have been published. Nagy et al. have extracted such objects and investigated them with the aid of an electron microprobe and found that some of these objects are fossilized with limonite probably and contain small amounts of nickel, while others are fossilized with silicates. Using hydrochloric and hydrofluoric acids they were able to remove these mineral constituents and show that there remained at least in some cases a body which could be seen under the microscope but which contained no elements heavier than magnesium, thus indicating that their composition was most probably that of carbonaceous material. This indicated that carbonaceous material had been fossilized by minerals much in the way that is observed in the case of terrestrial microfossils. Examples of their pictures are reproduced in Figs. 8 and 9.

To a chemist, morphological forms appear to be an unsatisfactory way of identifying biological remains because so many artifacts of approximately the size of microorganisms can be made, both from organic material and from inorganic material. However, it does seem reasonable to believe that specialists in the field of micropaleontology such as Staplin and Timofejev should be able to recognize common contaminants, and it is this author's reaction to take the work of reputable scientists in other fields as worthy

of serious consideration. It therefore seems to me that we should take these studies as not of negligible importance in connection with this problem.

Organic Chemical Studies

a. Fatty acids. More recently, studies using modern methods of organic geochemistry have revealed some very interesting chemical substances present in these carbonaceous chondrites. The Orgueil meteorite particularly has been studied in this way. Figure 10 shows infrared spectra taken on samples of naphthenic acid and extracts of brown alga, Orgueil meteorite and Bruderheim meteorite, and of the solvent blank. The material used for these spectra was extracted from the Orgueil meteorite or other material by benzene and methanol mixtures and saponified with potassium hydroxide, then extracted with water, acidified with HCl, re-extracted with ether, dried in nitrogen gas, and finally dissolved in carbon tetrachloride in which solvent the absorption spectrum was observed. It is evident that the extract from the Orgueil meteorite is very similar to what is observed from brown alga and naphthenic acid and that this material is not present in the Bruderheim meteorite nor in the solvent blank. The strong absorption peaks are characteristic of the carboxyl group. The marked similarity between these curves is indeed striking. The question immediately arises as to whether contamination of the Orgueil meteorite did not occur due to the growth of biological organisms which supplied the material observed in the test. Of course the Orgueil meteorite would have been biologically contaminated when it fell in France, but a museum shelf does not appear to be a particularly likely place for the growth of algae, particularly when the material contains solid magnesium sulfate which in the presence of a slight amount of adsorbed moisture would give a concentrated solution of magnesium sulfate. However, this possibility of contamination must be considered.

In Figure 11 are shown some chromatograms with a detailed caption. These are thin layer chromatograms on silica gel. The material was placed at the bottom of the plate and migrated upwards. The vertical columns 1 to 7 and 10 and 11 are from the Orgueil extract used to secure the infrared spectrum shown in Figure 10; 8 and 12 are prepared by the same procedure using petroleum naphthenic acids, and similarly 9 and 13 from an alga. Plate a was developed with normal hexane ether (97/3), b with the same solvents (95/5), and c with chloroform-methanol-water (65/25/4). The adsorbent was a silica gel layer and the components were made visible by spraying with Rhodamine 6G and by photography under ultraviolet light. The component A in Orgueil is saturated hydrocarbons, B is elementary sulfur, and C is the acidic compounds. This shows the absence of certain key biochemicals in Orgueil which are always present in terrestrial samples. Note the two light spots in 12 and 13 on plate c which are absent in Orgueil. These spots correspond to esters and sterols and they are present in the alga and naphthenic acid fractions but not in Orgueil. This study indicates that the simple supposition that an alga grew in the Orgueil rock on the museum shelf is not correct, or if such an alga did grow, it is a different one from those investigated in this work.

b. Optical activity. Optical activity of organic compounds has not been observed in nature or in artificial preparations and is characteristic of the action of biological organisms. Only the most careful and intricate technical processes have succeeded in separating the two optical isomers from the racemic mixture. The probability that this can occur in nature is exceedingly small and negligible, though it is conceivable that one optical isomer in a saturated solution could be seeded by a crystal of that variety and hence would separate out to a slight extent. The probability that this would occur in a chance stone such as a meteorite is very slight and can be neglected. Thus optical activity can be regarded as positive proof of the presence of biological material.

Figure 12 shows the results of measurements made by Nagy and his coworkers on the same extracts of the Orgueil material as used in the tests for the presence of carboxylic acids shown in Figure 10. The extraction procedure isolated lipids. The solutions are colored and hence only tests in the visible region can be made. But these tests were carried out in three different laboratories and by different investigators. One notes that the Orgueil meteorite is not similar in its optical activity to pollen grains, soil, or museum dust samples that were treated in the same way. Also, the blanks, though they scatter somewhat, seem to be definitely different from the Orgueil meteorite. Colored solutions in some instruments give a fictitious rotation and hence these authors used a blank containing sulfur and organic dye to imitate the color and opacity of the solutions being studied. The absorption spectrum of the Orgueil meteorite extract in this region and of the colored blank is shown on Figure 13. In this way an attempt was made to check on the possibility of error.

Recently, Hayatsu has used a somewhat different method of extraction and studied the optical activity. He claims to have secured negative results. Attempts by Nagy and his coworkers to reproduce Hayatsu's work have failed. They have not been able to remove sulfur from the solutions by the methods that Hayatsu describes, and they have found optical activity in their extracts at points where he finds none. Interestingly enough, they are able to get two fractions which show opposite optical activity. The extractions were made in their laboratories at the University of California, San Diego, and the tests for optical activity were made by Dr. W. D. Rosenfeld at the California Research Corporation, La Habra. It seems that complete objectivity in the tests could be expected under these conditions, and though the effects are small they nevertheless appear to be definite.

Table 3 briefly summarizes some of the results and the disagreement in this case. It may be that the difference between the two is due to some difference in the samples, i.e., it may be that Hayatsu was using a sample which did not contain biological material, while Nagy and his coworkers were using samples that had been contaminated by biological material. It is the writer's conclusion that optical activity is present in some samples. Contamination due to the growth of organisms after the meteorite arrived on earth is a possible explanation. Such organisms could have left residues of the compounds synthesized by them or could have consumed one optical isomer from an indigenous racemic mixture. There is some evidence that this is not the case but the subject must be pursued further in the future.

c. Porphyrins. Hodgson and Baker have published the results of a very detailed and careful study in regard to the possible existence of porphyrins in the Orgueil meteorite. The porphyrins have as their central chemical structure a grouping of four pyrrole rings linked together by four methine bridges (-CH-), forming a plane structure. The double bonds are such that long rings of conjugated double bonds are possible and several resonating structures exist thus contributing to the stability of the molecule. There are many points for ~~substitution~~ ^{substitution} of side chains on this central ring structure, and a divalent ion or radical occupies a position in the center, for example, Mg⁺⁺, Fe⁺⁺, VO⁺⁺, etc. Hemin which gives the red color to blood is a porphyrin and other biochemical compounds have this fundamental ring in their structure. On the other hand, chlorin is a name given to a group of compounds which differ from the porphyrin only in having one of the double bonds on one pyrrole ring hydrogenated with the addition of two hydrogen atoms. Chlorophyll has this central chlorin ring with characteristic side chains. The central ion in chlorophyll is magnesium. ~~Chlorophyll~~ Chlorophyll has a very characteristic

side chain, namely, one long alcohol group called phytol with 20 carbon atoms attached to a carboxylic group.

Studies on the distribution of porphyrins and chlorins in recent terrestrial deposits show that chlorins obviously obtained from chlorophyll mostly containing magnesium are prominent constituents. As the sediments become older, vanadyl and nickel porphyrins appear, indicating that the chlorins are oxidized by the removal of two hydrogens and the magnesium (Mg^{++}) is replaced by vanadyl (VO^{++}) and nickel (Ni^{++}). The ancient sediments and petroleum deposits contain only these porphyrins and both the vanadyl and nickel are prominent in these compounds.

The porphyrins have absorption bands in the neighborhood of 4000 angstroms. These are referred to as the Soret bands. They also have much weaker absorption bands at longer wavelengths as do the chlorins. The porphyrins are soluble in certain organic solvents and they can be separated from the hydrocarbons by solution techniques. The exact details of these processes are available in the literature of this subject and need not be repeated here.

Hodgson and Baker studied a number of samples of the Orgueil meteorite and two ordinary meteorites, Bruderheim and Peace River. Two Orgueil samples, one of 9 grams and another of 7 grams, were used especially for this study, while two other samples consisting of solutions which had been subjected to saponification procedures were also investigated for porphyrins. Briefly, it was found that the ordinary chondrites, Bruderheim and Peace River, showed no absorption bands in the region expected for porphyrins, that the 9 gram and 7 gram Orgueil samples showed a band at 412 microns which Hodgson and Baker interpret as due to vanadyl porphyrin, while the two samples which had been treated by saponification techniques showed no vanadyl porphyrin at all. But it was subsequently shown that the Posidonia shale from Germany, which is

known to contain vanadyl porphyrin, when treated by similar saponification processes also was depleted in its vanadyl porphyrin, thus confirming the observation that samples treated by the saponification technique previous to extraction with the organic solvents were depleted in vanadyl porphyrin.

It was the conclusion of Hodgson and Baker that "the Orgueil carbonaceous chondrite exhibits many of the organic components of ancient terrestrial rocks, and a detailed consideration of the environment of the Orgueil meteorite parent body by Nagy et al. led to the indication that the environment was a low temperature aqueous system with alkaline pH and slightly reducing redox potential. It is therefore not surprising to find what presently appear to be indigenous porphyrin pigments in the Orgueil stones suggesting a strong possibility of biogenic agencies in the origin of the organic matter of the meteorite." They found no evidence for nickel porphyrin which in view of its appearance in old sediments of the earth is surprising. Table 4 summarizes the features in the Orgueil extracts which lead to the identification of vanadyl porphyrin, and Figure 14 shows the absorption curves which they obtained. The didymium peak is for calibration purposes only. It will be noted that the shale and the Orgueil meteorite give curves which are very similar.

d. Nucleic acid bases. Calvin reported a cytosine-like feature in his investigation of the Orgueil meteorite. Oro has suggested that this may be an impurity, and as of the present this disagreement has not been satisfactorily solved. Hayatsu reported the presence of adenine and guanine and possibly a uracil type of compound. It should be noted that cytosine and uracil are the pyrimidine bases while adenine and guanine are the purine bases of ribonucleic acid. Hayatsu also reported other compounds that apparently have no biological significance and argues that for this reason these bases are

abiotic in origin. It should be noted on the other hand that many terrestrial deposits of organic material confidently believed to be of biological origin also contain compounds that have no known biological significance. Hence the presence of such compounds does not disprove the biological origin of other compounds.

e. Amino acids. Amino acids have been reported from the carbonaceous meteorites by Degens, Kaplan, Hamilton, and others. It appears certain that some of these are contaminations either by present-day organisms growing in the solutions being investigated or from human hands or things of this sort. The amount of the amino acids is very small and at present it is not possible to argue for an indigenous origin for these compounds, but it is also not possible to exclude that they at least partly have an indigenous origin. It should be noted that amino acids ^{may} decompose in terrestrial sediments more rapidly than the hydrocarbons, porphyrins, fatty acids, and other biological compounds of this kind.

f. Free radicals. Duchesne, Depireux, and Litt, using ^{nuclear} magnetic resonance techniques, have investigated the carbonaceous material of the Mighei and Nagoya (both Type 2) and report concentrations of free radicals up to 10^{17} per gram of carbon. They conclude that such quantities are characteristic of biogenic material. However, it is difficult to feel certain that non-biogenic carbonaceous material subjected to the radiations to which these meteorites were exposed would not imitate the free radical concentrations observed in terrestrial fossil carbonaceous material.

6. Hydrocarbons. Dr. Oro in his talk has referred to his very nice work on identifying hydrocarbons in a very considerable number of carbonaceous chondritic meteorites, and particularly the probable presence of pristane and phytane, two hydrocarbons generally believed to be degradation products of

the **phytole** side chain of chlorophyll. I only wish here to point out that his results and those presented in this review are consistent with each other. If these materials were of terrestrial origin, the suggestion would be made firmly that the materials were of biological origin and indigenous to the samples. This by all odds would be the most simple and direct interpretation of the results. A contrary explanation would be that some organism containing chlorophyll invaded these meteorites which have fallen all over the earth during more than a century in time, and which have been stored on dry museum shelves. These organisms containing chlorophyll grew vigorously for a short period of time and produced a record that in many ways duplicates what we find in very old sediments of the earth; at the same time they appear to lack important compounds such as that found in more recent soils and sediments of the earth. The former alternative would be generally accepted if these objects were of terrestrial origin, and in fact the second is so surprising as to be almost an unbelievable explanation.

Conclusions

Many lines of evidence, as briefly reviewed above, strongly suggest that biogenic material exists in these meteorites, and that it probably is indigenous. It seems safe to say that if this material were of terrestrial origin no question would be raised in regard to such a conclusion. Of course we know that life has evolved on earth, and hence the primary question and by all odds the most important question is decided for all samples of terrestrial origin. Certainly sufficient evidence has been found to justify very serious further work on these objects, and I am sure that this will be done by several groups in the future.

Possible Origins of Fossil-Bearing Meteorites

Nagy, Meinschein, and Hennessy's paper in 1961 aroused great opposition on the part of students of meteorites because of the chemical composition of these objects. Those of us who have been working on meteorites for some years

were certain that there could not be the residue of living things in these objects. Had the meteorites had the composition of sedimentary rocks on the earth, no great surprise would have been expressed. It would have been assumed that they came from some planet, probably in the asteroidal belt, which had existed at some time in the past and had been broken up by some process. The fact that the carbonaceous chondrites have a chemical composition with respect to the usual non-volatile fraction of primary matter, namely, the silicates and related compounds, which is approximately that of the sun, made it exceedingly difficult to understand how biological material could have originated in these objects.

We are questioning whether the carbonaceous chondrites have life very closely related to terrestrial life, and such life as we know it exists only in water. Also, the chemical experimentation required to evolve the complicated and intricate processes of living things requires a source of free energy, for only such free energy makes this chemical experimentation possible. Hence the surface of a planet large enough to retain water at a temperature such that liquid water would exist on its surface, and the presence of solar radiation, provide the only possible conditions for the evolution of life. If this planetary object was partially covered with water, then oceans, rivers, and all the erosional processes of a planet such as the earth would be present. Sedimentary rocks should have been formed and such sedimentary material should be arriving at the earth as meteorites. This is definitely not the case. No observed case of a sedimentary meteorite has ever been established. It should be noted that all the carbonaceous chondrites are observed falls. If these can be observed to fall, then any possible sedimentary meteorites could also have been observed.

The other possibility is that the planet was completely covered with water. In this case no sedimentary rocks would be expected, but also the biological remains would be deposited only in thin layers at the bottoms of the oceans. In this case the probability of securing a sample of this material would be very small and as noted above the carbonaceous chondrites make up approximately three per cent of the total observed falls. Hence this assumption is impossible.

There is left only the possibility that life evolved on one planetary object and that it was transferred to another planetary object of primitive composition. Of course the example of this in the solar system that immediately comes to mind is the earth, where we know life has evolved, and the nearby moon which may have a composition consistent with the carbonaceous chondrites for all we know. It is an old suggestion that the meteorites have been coming from the moon, but the recent evidence, though not conclusive, is suggestive at least.

If the moon escaped from the earth it is not unreasonable at all to believe that it could have been contaminated with terrestrial water temporarily. If the moon was captured by the earth, the process may have been a very complicated and violent one, because such a capture hypothesis almost surely implies that many moon-like objects were about and that they and fragments of them were accumulated into the earth, a hypothesis that was put forward by Urey some time ago. If indeed the surface of the moon carries a residue of the ancient oceans of the earth at about the time that life was evolving, it means that the Apollo Program should bring back to us fascinating samples which will teach us much in regard to the early history of the solar system, and in particular in regard to the origin of life.

Some people are exceedingly skeptical in regard to the interpretation of life in these objects and probably the great importance of the subject justifies skepticism. On the other hand, we in my own country and I believe it is also true of the U.S.S.R., are making extensive plans to explore the solar system, and one of the most fascinating results of all this exploration would be the proof that life may exist somewhere else than on earth. It seems that we should be willing to consider the evidence for a residue of life in meteorites objectively, and that we should not draw final negative conclusions before all the evidence has been secured. Moreover, some tolerance for those of us who hope that the residue of life exists in these objects would be consistent with great enthusiasm for exploring space.

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6. Free Radicals.

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FIGURE CAPTIONS

1. Petrographic thin-section of the Orgueil meteorite. Petrographic thin-sections are prepared by the careful grinding of a meteorite fragment cemented to a glass microscope slide with a suitable plastic. This photomicrograph shows the fragmental (brecciated) texture of Orgueil (note granule at upper left corner) and a magnesium sulfate vein (in vertical position, just to the left of center). Such textural features are not present in unaltered igneous rocks. The mineral matrix, with the exception of opaque granules and the magnesium sulfate vein, consists of clays. The horizontal distance between the two edges of this photomicrograph is 0.7 mm.
2. Another petrographic thin-section of Orgueil. This one shows preferred orientation of the opaque lath-like minerals. This textural pattern is probably the result of plastic flow of the clay matrix (not the result of the grinding of the thin-sections) on the parent body. Such localized flow structures are characteristic of pyroclastic sediments (such as volcanic tuffs). Pyroclastic sediments are the agglomerated debris of broken up igneous rock bodies.
3. Acid insoluble residues of organized elements from Orgueil. (After Staplin) (HF was used to remove silicates and this was followed by treatment with a mixture of conc. HNO_3 and KClO_3).
4. Acid insoluble residues of organized elements from the Mighei meteorite. (After Timofejev) The mineral matter was removed with conc. HNO_3 and potassium permanganate solutions. The organic residues were concentrated by density separation during centrifugation.

FIGURE CAPTIONS (Cont'd.)

5. Petrographic thin-section of Orgueil showing an organized element embedded in the mineral matrix.
6. Photomicrographs of an organized element from Orgueil (treated with HCl and dyed with fuchsin), shown left, and of the organic shell of the fossil marine alga Xanthidia from Tertiary rocks in Australia, shown right. The alga was stained with safranin and it is 35-40 μ in diameter. It is reproduced from a paper by W.A.S. Sarjeant of Nottingham University. The visible ultramicrospectrum of the Orgueil organized element, measured at the Karolinska Institute last September, was wholly different from identically treated recent biological matter, such as ragweed pollen.
7. Organized elements (untreated) in a powdered preparation of Orgueil.
8. Orgueil organized elements (photomicrographs and the corresponding drawings), analyzed by the electron microprobe. 1-6 and 9 contained only Fe with traces of Cl and/or Ni. 7 and 8 were iron-magnesium silicates. This particular microprobe could not detect elements below Mg in the periodic table.
9. The lack of the back-scattered electron image of the electron microprobe, shown on the right, of an organized element from Orgueil which was freed from mineral matter by boiling it in 6N HCl for one hour. Photomicrograph of the particle is shown on the left and a drawing of it in the center. The lack of the back-scattered electron image of this acid insoluble residue points out that elements heavier than Na were absent, and it suggests that the residue is composed of organic matter.

FIGURE CAPTIONS (Cont'd.)

10. Infrared spectra of solvent soluble, saponifiable organic matter extracted from Orgueil and from controls. Note that the procedure blank shows that no contamination occurred in the laboratory. The peaks at 5.85 and 10.7 μ and the smaller absorption peaks at 3.3 and 3.7 μ are characteristic of carboxylic acids (such as fatty and aromatic acids). All samples shown on this slide were extracted for six hours in benzene-methanol, saponified with KOH, extracted with water, acidified with HCl, re-extracted with ether, dried under N₂ and dissolved in CCl₄.

11. Thin layer chromatograms of the Orgueil extract, the infrared spectra of which were shown in the previous slide. The vertical columns (i.e., applications) 1-7 and 10,11 are from the same Orgueil extract, 8 and 12 are identically prepared extracts from petroleum naphthenic acids, and 9 and 13 from an alga. Plate a was developed with n-hexane-ether (97:3, v/v), b with the same solvents (95:5, v/v), and plate c with chloroform-methanol-water (65:25:4, v/v/v). The adsorbent was a silica gel layer and the components were made visible by spraying with Rhodamine 6G and by photography under ultraviolet light. The component A in Orgueil is saturated hydrocarbons, B is elementary sulfur, and C is the acidic compounds. This slide is important because it shows the absence of certain key biochemicals in Orgueil, which are always present in terrestrial samples. Note the two light spots (in 12 and 13), below the middle of plate c which are absent in Orgueil. These spots correspond to esters and sterols and they are present in the alga and naphthenic acid fractions (12 and 13) but not in Orgueil (10,11).

FIGURE CAPTIONS (Cont'd.)

12. Optical activity of the same Orgueil extract which was shown on the two previous slides. Two other Orgueil extracts, prepared identically but from two other stones, are also included. The measurements were made at three different laboratories by three independent investigators. Range of instrumental error is shown by vertical lines. α is the observed rotations. The specific rotation is approximately 1° .
13. The visible spectra of the optically active Orgueil extract and of a blank containing both colloidal sulfur and a synthetic dye. The strongest optical activity in Orgueil was measured at the wavelength where the arrow is located. The blank, which has the same absorbance at this wavelength, showed no optical activity.
14. Spectra of a chromatographic fraction of Orgueil showing absorption bands characteristic of vanadyl porphyrins. Note that the Orgueil band intensity is approximately the same as that of the Posidonia shale from Germany, which is a classical source of ancient porphyrins. Didymium is a wavelength calibration standard.

TABLE CAPTIONS

1. Elementary compositions (atomic percentages with S, C, and H₂O excluded) of two carbonaceous meteorites and the average composition of the H-group chondrites. The carbonaceous chondrites show close agreement (with the exception of metallic Fe and iron oxides and sulfides) with the non-carbonaceous chondrites.
2. The sulfur, water, and carbonaceous matter content of carbonaceous meteorites and of the H-group chondrites.
3. Optical activity of Orgueil extracts, prepared by Nagy using the method of Hayatsu, compared with those reported by him.
4. Summary of additional experiments (extractability, chromatographic behaviour, spectral band shift with different solvents, chemical reactions) which point out that the Orgueil fraction, shown in Fig. 14, contains ancient porphyrins.

Table 1

	Type I Orgueil	Type II Meghei	Average H.Chondrites	
Fe (Metal)	0.00	0.00	16.18	
Fe (Oxide- Sulfide)	27.34	26.18	10.05	
Ni	1.37	1.41	1.57	
CO	0.07	0.06	0.09	
Si	31.12	31.85	33.12	Atomic percentages S, C, water eliminated
Ti	0.09	0.09	0.09	
Al	2.68	2.90	3.60	
Mn	0.22	0.19	0.25	
Mg	32.48	33.19	31.68	
Ca	1.81	2.04	1.60	
Na	1.97	1.40	1.56	
K	0.12	0.07	0.19	
P	0.33	0.29	0.19	
Cr	0.40	0.33	0.33	
Total	100.00	100.00	100.00	

Table 2

	Type I Orgueil	Type II Meghei	Average H.Chondrites	
S	5.50	3.66	1.57	Wt. percentages
H ₂ O	19.89	12.86	0.37	
Carbonaceous	6.96	2.48	----	

Table 3

OPTICAL ACTIVITY MEASUREMENTS
ON ORGUEIL EXTRACTS PREPARED
BY THE METHOD OF HAYATSU

Measurements were made by the California Research Corporation on samples prepared at the University of California, San Diego.

$$\text{Error} = \pm 0.005^\circ$$

Measured at 546 m μ

Sample	Hayatsu	Univ. of California, SD	1,2
Saponifiable, (A ₃)	-0.001	-0.025	
Non-saponifiable, (A ₂)	-0.001	+0.014	

1. A colored blank prepared by refluxing sulfure with benzene-methanol, as described by Hayatsu, showed no rotation at this wavelength (-0.005)
2. All San Diego results were corrected for complete process blanks.

Table 4

EVIDENCE FOR PORPHYRINS IN THE ORGUEIL METEORITE

Main Spectral Absorption Features in the Orgueil Extracts

387-388 m μ — not porphyrin
410-412 m μ — probably porphyrin
430-445 m μ — not porphyrin

Examination of 410-412 m μ pigment

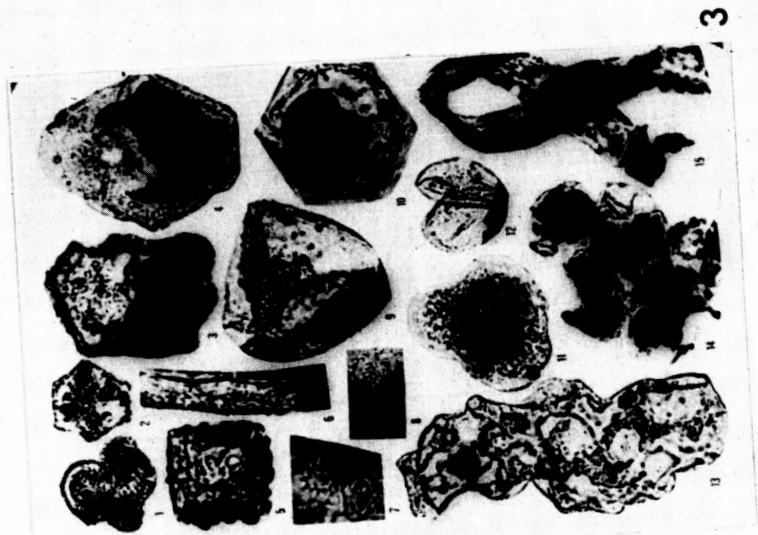
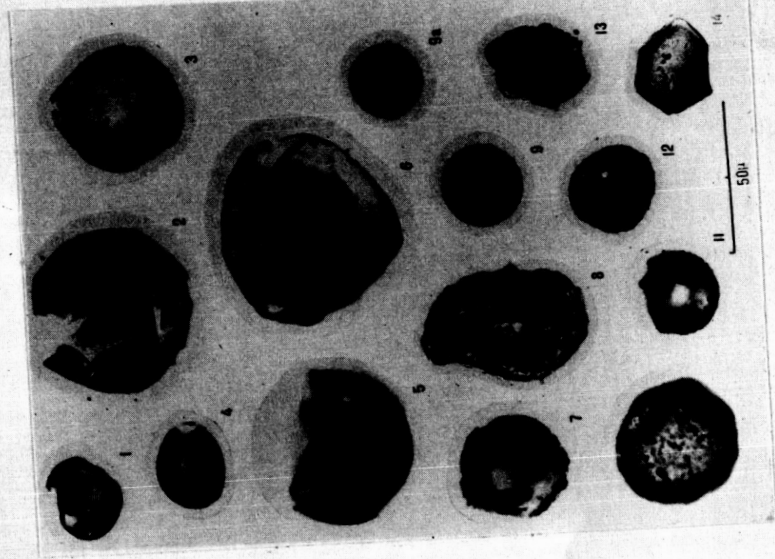
Extractability from carbonaceous rock — similar to terrestrial sediment porphyrins
Chromatography on silica gel — similar to terrestrial sediment porphyrins
Spectral shift of Soret band with solvent — similar to terrestrial sediment porphyrins
Chemical Reactions
—HBr—HOAc decompositions (limited test) — similar to terrestrial sediment porphyrins
—KOH and sulphur decomposition — similar to terrestrial sediment porphyrins

Origin of 410-412 m μ pigment

Contamination by dusts, soils and recent sediments — unlikely, due to virtual absence of chlorins
Indigenous — likely

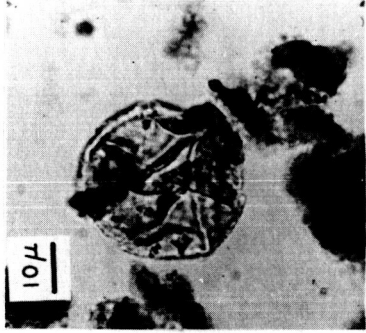
Probable identity and concentration of 410-412 m μ pigment

Esterified vanadyl porphyrin — 0.01 p.p.m. of meteorite

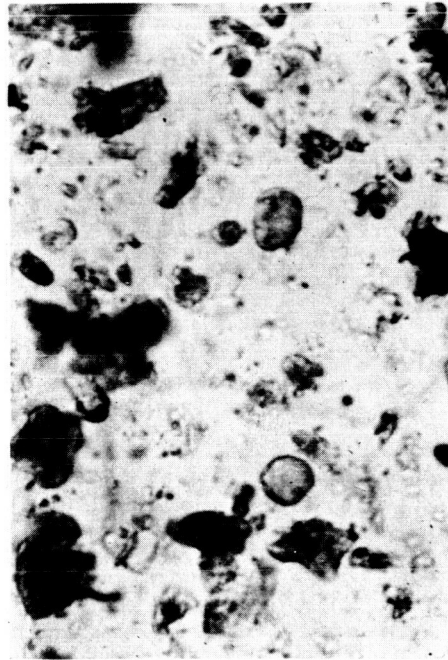




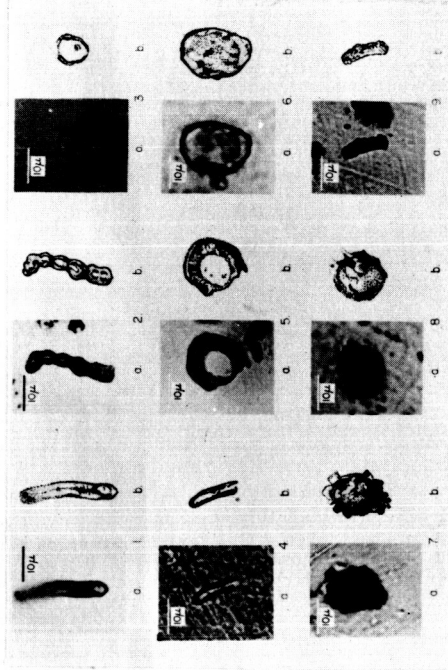
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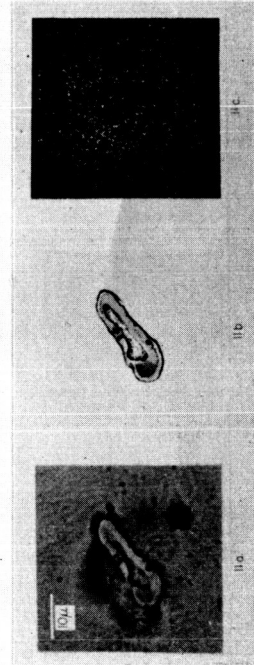


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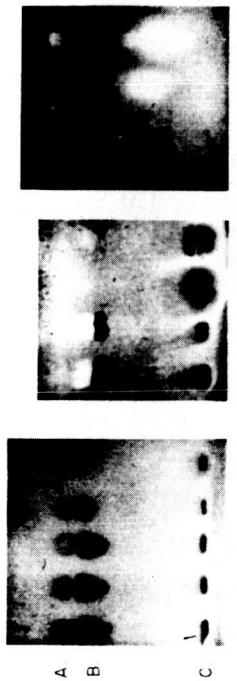
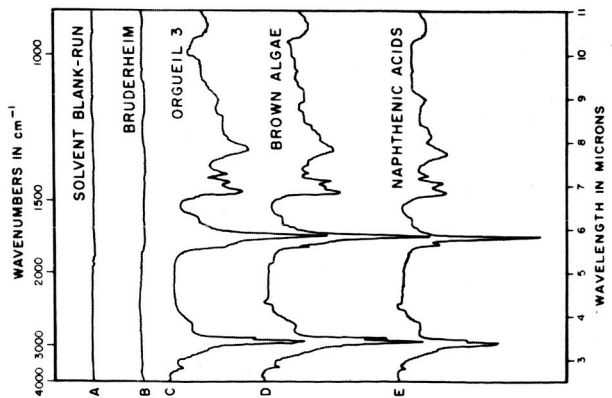


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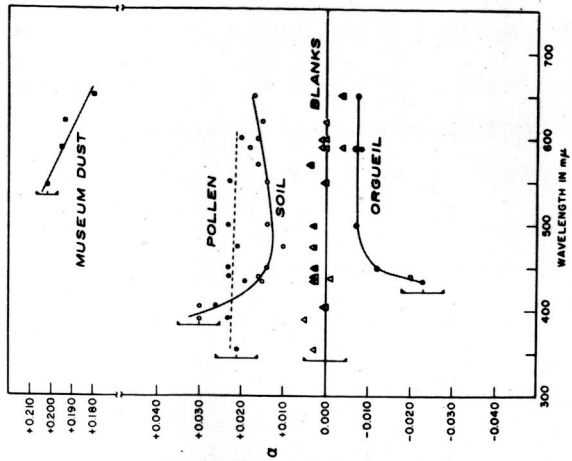


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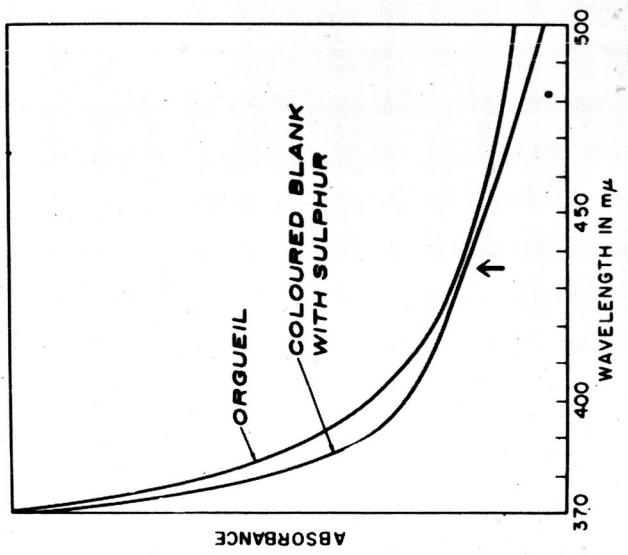


a. b. c.

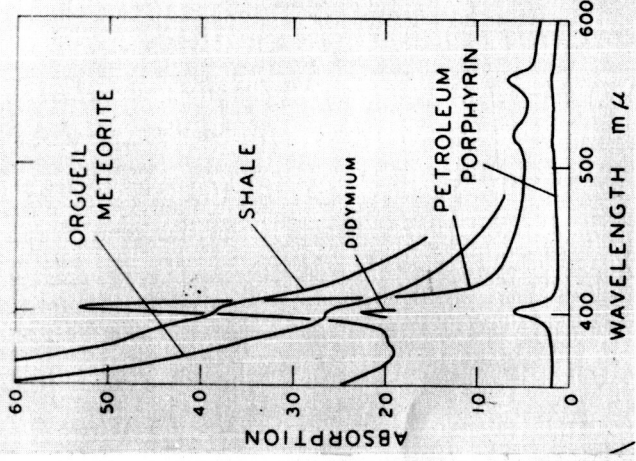
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12



13



14