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Organic Compounds in Carbonaceous Chondrites

These compounds seem to have formed in the solar nebula by equilibrium reactions among hot gases.

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How did the organic compounds in meteorites form? Were they made by living organisms in outer space, or by purely chemical, abiotic processes?

No consensus has been reached on these questions, although they have been debated for more than a century. Most scientists nowadays believe that these compounds formed abiotically. But their formation conditions have remained in doubt, for lack of definite clues. We believe such clues have been obtained from the traces of volatiles trapped in these meteorites.

All carbonaceous chondrites contain traces of primordial noble gases, firmly held within mineral grains. These gases were presumably trapped from a primitive gas phase (atmosphere or nebula) during the formation of the carbonaceous chondrites or their minerals. To explore the possibility that other volatile constituents of the gas phase were trapped at the same time (*1*), we studied three carbonaceous chondrites by time-of-flight mass spectrometry.

Procedure

A sample containing a few milligrams of carbonaceous chondrite was placed in a copper side arm of an all-metal vacuum system which terminated in the source region of a modified Bendix time-of-flight mass spectrometer (*2*). The sample tube was cooled with liquid nitrogen and evacuated with a system separate from that of the spectrometer. The liquid nitrogen was removed from the sample, and the evacuation was continued through a U-tube cooled with liquid nitrogen. After N_2, O_2, and Ar had been removed, the system was closed and any gases evolved were permitted to collect over the sample at room temperature. Periodically the sample tube was closed off and the collected gases were examined by the momentary cracking open of a valve to the spectrometer. The spectrum to mass 500 was observed, and regions of interest were expanded and photographed.

After the uncondensed gases had been examined and pumped away, the auxiliary vacuum system was closed off. The liquid N_2 was removed from the U-tube and the condensed vapors were allowed to distill into a part of the closed system which was cooled with liquid nitrogen. During the transfer the valve into the spectrometer was opened periodically and the distilling species were examined. Effective separations of many-component systems were often obtained. The entire distillate collected up to this point was then warmed and pumped away, and the process was repeated. The temperature of outgassing was gradually raised, and samples were collected for periods of up to 16 hours. Results obtained at temperatures above 140°C were discounted because of possible thermal degradation or other chemical reactions.

In one series of experiments, designed to sort out possible terrestrial contaminants, chunks of meteorite, after preliminary degassing at 100°C, were broken up under vacuum by the application of a vibrating engraving tool to the outside of the sample tube. Both H_2 and CH_4 were then seen to evolve at room temperature. Fresh quantities of the less volatile and more tightly held constituents were evolved at higher temperatures. Both CO and N_2 were observed, without O_2.

Compounds of low abundance or low volatility were sought in solvent extracts of much larger samples of meteorites (5 to 20 grams). In the larger samples, these compounds were more readily detectable.

Compounds were identified from the observed spectra by comparison with fragmentation patterns published by the American Society for Testing and Materials and the American Petroleum Institute.

Results

Three carbonaceous chondrites (Orgueil, Cold Bokkeveld, and Murray) and six oil shales were studied (*3*). The substances identified in the meteorites are listed in Table 1, in order of increasing molecular weight.

The prominence of aromatic hydrocarbons, as shown in Table 1, is remarkable (*4*). Aliphatic hydrocarbons, on the other hand, were present only in subordinate amounts in the three meteorites. A striking and exceedingly important finding is the scarcity of ethane relative to methane. An upper limit to its abundance is $\leq 10^{-3}$ the amount of methane.

The apparent absence of NH_3 is also interesting, since NH_3, rather than N_2, has always been assumed to be the

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principal form of nitrogen in primitive atmospheres. Ammonium ion, likewise, has not been found in carbonaceous chondrites (*5*). The observation of chlorinated hydrocarbons confirms early work by Mueller *6*).

Sulfonic acid esters have not been previously reported from meteorites. A definitive identification was made from the infrared spectrum. From the vantage point of hindsight, their presence is not completely surprising, since the meteorites contain large amounts of magnesium sulfate (*5*).

Table 1. Volatiles in carbonaceous chondrites. Approximate relative abundances are indicated by one or two plus signs; compounds sought but not found, by minus signs; compounds seen in solvent extracts, by E. Question marks indicate tentative identifications. The data represent 28 separate analyses.

Compound	Orgueil	Cold Bok-keveld	Murray
H_2	++	+	++
He			+
CH_4	++	++	++
NH_3	—	—	—
H_2O	++	++	++
CO	++		++
N_2	++	++	++
NO	++	++	+
C_2H_6	—	—	—
Ar^{40}		++*	
C_3H_8	—	—	—
CO_2	++	+	
C_4H_8	++	++	++
COS	++	++	
SO_2	++		
CS_2	++		++
C_6H_6	++	++	++
Kr	++		
$C_6H_5CH_3$	++	+	
$C_6H_5C_2H_5$	+		
$C_6H_4(CH_3)_2$	+E		
C_6H_5Cl	+		
$C_6H_5C_3H_7$	+		
$C_6H_5[CH(CH_3)_2]$	+		
$C_6H_3(CH_3)_2(C_2H_5)$		++	
$C_6H_4Cl_2$		+	
Xe	++		
$C_{10}H_8$	+E		
$C_6H_4(C_2H_5)_2$	+E		
Anthracene Phenanthrene	E		
$C_nH_{2n+1}Cl$ ($n = 12$ to 18)	+	+	
$R-SO_3-CH_3$†	+E		
Trimethylanthracene or trimethyl-phenanthrene	E		
Alcohols	?	?	+
Aliphatic and alicyclic hydrocarbons	?	?	

* At 190°C. † Each of three peaks, at $m/e \approx$ 207, 281, and 355, was accompanied by two satellite peaks, 16 and 32 mass units lower. The infrared spectrum strongly suggests that these compounds are sulfonic acid esters. Owing to the strong fragmentation tendency of sulfonic acid esters, it is possible that the parent molecules have masses appreciably higher than 207, 281, and 355.

Prominent unidentified peaks were seen at the following mass-to-charge (*m/e*) ratios: Orgueil A, 130, 132; Orgueil B, 71, 72, 96, 113, 120, 121, 135, 170, 191; Orgueil C, 58, 69, 70, 71, 72, 95, 96, 97, 110, 112, 113, 115, 132, 134, 136, 138; Cold Bokkeveld, 56, 81, 96; Murray, 58, 69, 70, 71, 96.

In comparing the relative abundances of the volatiles, it must be borne in mind that the meteorites had been heated to ~300°K at every perihelion passage during the last few million years of their history. Highly volatile compounds therefore survived only in a few retentive sites inside mineral grains, while less volatile ones were retained in these sites as well as in surface coatings and interstitial material.

The H_2 and CH_4 were definitely indigenous, not contaminants or degradation products. An unground Orgueil sample and a fine powder of Cold Bokkeveld prepared 10 years earlier gave off no methane and very little H_2 even when heated to 140°C. Freshly ground samples of all three meteorites yielded H_2 and methane at room temperatures. Thus it is unlikely that the H_2 and the CH_4 could have been produced by thermal degradation of more complex materials. They appeared in the least condensable fractions, where no higher-molecular-weight organics were present. Hence they could not have been produced by thermal cracking processes in the mass spectrometer.

Some small fraction of the volatiles must, however, have been made by a preterrestrial degradation process—by action of cosmic rays and of natural radioactivities on complex organics. The total radiation doses from these two sources are, respectively, about 100 and 500 megarads (for a cosmic-ray age of 3.5 million years, as for Orgueil, and for chondritic levels of potassium, uranium, and thorium). Hence, about 1 percent of the original bonds might have been ruptured by radiation damage (*7*). That this process played only a minor role is shown by the absence of ethane, propane, and their heavier homologs in the meteorites. Radiolysis of complex organic materials, be they synthetic polymers or natural hydrocarbons, invariably produces comparable amounts of methane and ethane (*8*). As a control, we put six samples of Green River and Chattanooga oil shales through our analytical procedure. The radiation doses for these two shales seem to have been about 300 and 3000 megarads, respectively.

The results confirm previous studies by Hoering and Abelson (*9*). The four shales which have a firm enough texture to retain methane display a continuum of lighter aliphatic hydrocarbons, with methane and ethane present in comparable amounts. In the meteorites, ethane and propane are completely lacking; since methane is present, the absence of its higher homologs cannot be blamed on losses from reheating or diffusion.

The oil shales provided a good check on the technique. As the condensate was warmed from −195°C to room temperature, H_2 and, successively, aliphatic hydrocarbons from C_1 to C_{20} appeared in the spectrum.

Equilibrium or Nonequilibrium Processes?

If we accept the view that the volatiles in carbonaceous chondrites are relatively unaltered relics of the primordial gas phase, we can attempt to draw inferences about the nature of this gas phase. Two principal alternatives for the origin of the organic compounds must be considered.

1) High-energy, nonequilibrium processes—for example, Miller-Urey reactions involving ultraviolet or charged-particle irradiation. These processes are usually assumed to have taken place in a reducing atmosphere of CH_4, NH_3, and H_2O, since Urey (*10*) has shown that carbon, nitrogen, and oxygen would be present in the form of their hydrides in a cold nebula of cosmic composition.

2) Equilibrium processes—that is, reactions among the constituents of the gas phase under conditions of thermodynamic equilibrium.

The concept of equilibrium processes has been in limbo for the past 12 years, partly because thermodynamic calculations showed them to be incapable of producing more than traces of complex organic compounds in a gas phase of cosmic composition, and partly because Miller-Urey reactions were so astonishingly successful in synthesizing such compounds. The first attempt to break this spell was made by Suess (*11*), who pointed out that complex organic compounds might form by equilibrium reactions in a *fractionated* gas phase, in which the hydrogen-carbon ratio had fallen to about 1/1000 its cosmic value. This idea was pursued further by Dayhoff, Lippincott, and Eck (*12*), who computed equilibrium concentrations of over 100 organic compounds for various proportions of carbon, hydrogen, oxygen, and nitrogen. Temperatures and pressures ranged between the limits 300° and 1000°K and 10^{-6} and 300 atmospheres.

Dayhoff's calculations were truly representative of equilibrium processes in general, covering as they did all principal regions of the C-H-O diagram (Fig. 1). This suggested a possible test of the alternatives under discussion. If the organic compounds in meteorites had formed by equilibrium processes, the observed distribution should match one of the calculated distributions more or less closely. A complete mismatch would be equally informative, since Dayhoff's calculations seem to have covered all principal possibilities.

An important feature discovered by Dayhoff and her co-workers was the existence of an "asphalt threshold" in the phase diagram. On the carbon-poor side of this line (for example, points *A*, *B*, and *D* in Fig. 1), only traces of hydrocarbons and other organics form. On the carbon-rich side (for example, point *C*), aliphatic hydrocarbons are still quite scarce but the abundance of aromatic hydrocarbons is enhanced by factors of 10^{20} to 10^{30}, and many other organics are also produced in increased yields. Nearly one-tenth of the total carbon is converted to complex, polynuclear aromatics, described by the collective term "asphalt."

In Table 2 the calculated distribution of volatiles in the asphalt region (distribution C of Dayhoff *et al.*) is compared with the observed distribution in carbonaceous chondrites. In the calculated distribution, we have listed only compounds with abundances greater than 10^{-10} mole per mole of total carbon, ranked according to abundance.

The similarity between the calculated distribution C and the observed distribution in meteorites is remarkable. The nine compounds listed in distribution C as most abundant are present in carbonaceous chondrites. The next two compounds in order of distribution-C abundance, ethane and ammonia, were not detected with certainty, in spite of careful searches. The apparent level of ethane, $\leq 10^{-3}$ the abundance of methane, is higher than the calculated abundance by a factor of 10. Of the last five compounds of distribution C, only oxylene was found, favored apparently by higher sensitivity of detection in the region near mass 106. Three of the remaining four compounds have low molecular weights and are apparently too rare to be detected by direct volatilization and too volatile to be recovered by solvent extraction. Thus there is a striking parallelism between the calculated and the observed distributions. Compounds predicted to be abundant are indeed abundant, and compounds predicted to be rare are undetectable. None of Dayhoff's other distributions give a satisfactory match.

When these comparisons are extended to various distributions observed in nature or in the laboratory (most of which have originated under nonequilibrium conditions), it soon becomes apparent that both the meteorite distribution and distribution C are well-nigh unique. One feature that sets them apart from most alternative distributions is the absence of the heavier homologs of methane. Degradation of biogenic or abiogenic hydrocarbons by heat or radiation invariably produces a continuum of lighter aliphatic hydrocarbons (*8*, *9*), and so does the radiation-induced polymerization of methane (*13*). Another characteristic feature is the overwhelming preponderance of aromatic over aliphatic hydrocarbons. Vinogradov and Vdovykin (*14*) have recently shown that even the insoluble carbonaceous matter in carbonaceous chondrites, comprising some 50 to 90 percent of the total carbon (*15*), is an aromatic polymer. While some of the processes cited above can convert aliphatic hydrocarbons to aromatic hydrocarbons, the homologs of methane are always produced in such reactions. Equilibrium reactions in the asphalt region seem to be the only ones that can reconcile the preponderance of aromatics with the lack of ethane and its homologs.

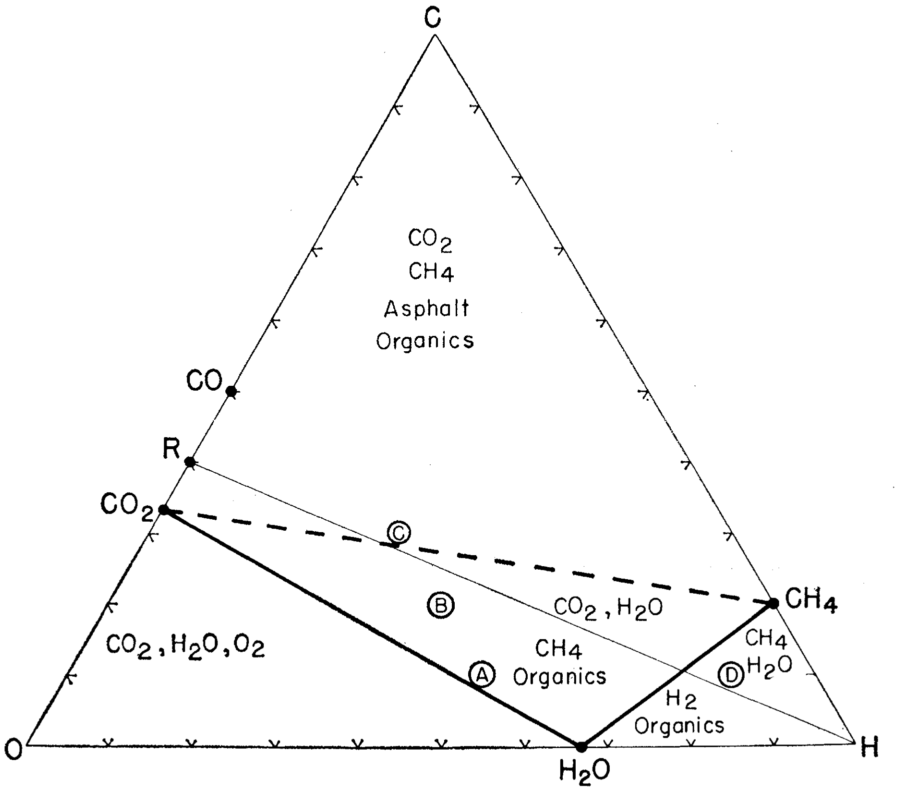

Fig. 1. Carbon-hydrogen-oxygen equilibrium diagram, indicating principal compounds expected in each field (*12*). Points *A–D* correspond to detailed product distributions computed by Dayhoff *et al.* Large amounts of aromatic compounds, such as are found in meteorites, are formed only in the "asphalt" region in the upper part of the diagram. The lower boundary of the asphalt region at 500°K is given by the dashed line. The boundary is slightly higher at higher temperatures. A gas of cosmic composition (initially near *H*) can move into the asphalt region by simple loss of molecular hydrogen as represented by the compositional track *HR* (*R* corresponds to the cosmic carbon-oxygen ratio).

Perhaps, then, this is the principal mechanism by which the organic compounds in meteorites formed.

Two further arguments tend to support this suggestion. First, the organic compounds in Orgueil show an appreciable spread in C^{13} content, up to 15 per mil (*16*). High-energy, nonequilibrium processes, such as radiolysis and Miller-Urey reactions, are not likely to lead to appreciable isotopic fractionations, whereas equilibrium processes are. Second, a study of the isotopic composition of sulfur, $MgSO_4$, and FeS in the Orgueil meteorite (*17*) shows that elemental sulfur was the precursor of the other two oxidation states. But the stable form of sulfur in a gas of cosmic composition is H_2S at high temperatures and FeS at low temperatures (*18*). If elemental sulfur formed instead, the gas phase must have been greatly depleted of hydrogen.

The statement that the organic compounds formed under equilibrium conditions must be qualified in one important respect. Strictly speaking, elemental carbon is the stable product in the asphalt region. Dayhoff and her co-workers assume that the formation of carbon was inhibited by kinetic factors, so that organic compounds formed instead, as metastable products. As far as the meteorites are concerned, this is not an unreasonable assumption. They contain organics, but no free carbon; hence the formation of elemental carbon must indeed have been inhibited. The implied short reaction times are not unprecedented. Certain meteoritic features, such as chondrules, seem to have formed on a time scale of minutes or less (*18*).

Metastable formation in partially equilibrated mixture may also account for certain compounds, such as amino acids, nitrogen bases, and saturated hydrocarbons, which seem to be more abundant in meteorites than is predicted by Dayhoff's calculations (*19*). Alternatively, these compounds may have been made by special processes, such as Miller-Urey reactions or hydrogenation of aromatic hydrocarbons.

The evidence, then, suggests rather strongly that the organic compounds in carbonaceous chondrites formed largely under conditions of thermodynamic equilibrium in a primitive gas phase. But can anything more definite be said about this gas phase? When and where did it exist in the solar system?

Time and Place of Formation

There is scarcely any doubt that most or all meteorites come from the asteroids, bodies that were never larger than a few hundred kilometers in diameter (*20*). For a rare class, such as the carbonaceous chondrites, it is difficult to exclude an origin from an alternative source—for example, comets (*21*). For cometary or asteroidal material, three types of gas phase must be considered: (i) a temporary atmosphere; (ii) an "internal" atmosphere of gases escaping from the hot interior of an asteroid; (iii) the solar nebula itself.

The first two possibilities can probably be excluded. For one thing, an impossibly dense atmosphere (100 to 1000 kg of $CO_2 + CH_4$ per square centimeter) would be required to account for the observed amounts of organic matter in meteorites (~0.05 percent by weight). Moreover, some selective loss of CH_4 relative to H_2O would be bound to occur during the accretion and subsequent heating-up of the body. With a cosmic carbon-oxygen ratio of 0.67 (*22*), even a slight preferential loss of methane would move the system below the threshold of the asphalt region ($C/O = 0.50$).

This leaves the solar nebula as the principal possibility. Initially, the nebula must have had "cosmic" composition (a composition nearly coinciding with point *H* in Fig. 1), with hydrogen, carbon, and oxygen largely in the form of H_2, CH_4, and H_2O. The simplest process that would bring this mixture into the asphalt region involves selective loss of H_2, with retention of H_2O and CH_4. This path is represented in Fig. 1 by the straight line joining *H* to *R* (*R* is the cosmic C/O ratio). Alternative paths, involving concurrent loss of CH_4 and H_2O, can be ruled out. In any such fractionation process, CH_4 would be lost preferentially, owing to its lower molecular weight and boiling point. The compositional track would thus veer downward from the line *HR*, missing the asphalt region entirely.

Table 2. Calculated and observed concentrations of volatiles.

Compound	Calculated distribution C*	Observed in carbonaceous chondrites†
Carbon dioxide	0.66	+
Nitrogen	.50	+
Methane	.24	+
Carbon monoxide	5.3×10^{-3}	+
Benzene	6.6×10^{-4}	+
Naphthalene	1.4×10^{-4}	+
Hydrogen	1.3×10^{-4}	+
Water	7.9×10^{-5}	+
Anthracene	2.9×10^{-5}	+
Ethane	2.3×10^{-5}	–
Ammonia	1.1×10^{-6}	–
Xylene	9.4×10^{-8}	+
Ethylene	5.7×10^{-9}	
Acetic acid	1.2×10^{-9}	
Hydrogen cyanide	1.4×10^{-10}	
Benzoic acid	1.3×10^{-10}	

* Equilibrium concentrations, in moles per mole of total carbon at 500°K and 1 atm. Initial C : H : O ratio = 1 : 1 : 1.33 (see *12*). † Plus sign, observed; minus sign, sought but not found.

It is difficult to decide whether such a selective loss of H_2 is a reasonable suggestion. Isotopic data on sulfur in Orgueil suggest a hydrogen-poor environment prior to accretion (*17*). The depletion factors for primordial neon, argon, krypton, and xenon in primitive meteorites are a smooth function of mass number (*20*), a finding which, like other considerations, points to a mass-dependent fractionation process. Cameron's model for the formation of the solar system demands, on the other hand, that the gases were lost in bulk, without fractionation (*23*).

An important clue is the intimate association of the volatiles and the mineral grains. The thermal-release patterns of the volatiles which we studied and of the noble gases (*24*) suggest that the molecules are not superficially adsorbed but are firmly held within mineral grains. It seems difficult to avoid the conclusion that the mineral grains were formed in a gas phase containing the volatiles.

The following tentative picture can be pieced together from these considerations. At some stage in the formation of the solar system the inner part of the solar nebula underwent a mass-dependent fractionation process, which removed hydrogen and helium, at least on a local scale. The C:H:O ratio of the residual dust and gas lay in the asphalt region. Part or all of the material was then heated and vaporized. When this vapor cooled, a broad spectrum of organics was produced, by equilibrium reactions and by some metastable reactions involving ammonia. Mineral grains continued to grow from the supersaturated-gas phase (*18, 25*), trapping small amounts of noble gases and other volatiles in the interior of their crystal lattices. Major amounts of ices and less-volatile organics condensed directly, or on the surfaces of mineral grains. The dust grains and any residual gas were irradiated by means of charged particles from the sun (*20, 26*). This irradiation, and perhaps also reactions with hydrogen, led to the formation of a second, smaller crop of organics. Ultimately, the dust grains accreted to form planets, asteroids, and comets. In small asteroids or in the outer zones of larger asteroids the dust suffered little change, except for alteration by the action of liquid water (*4, 5*) and a slight degree of thermal metamorphism (*18*). The carbonaceous chondrites presumably come from such locales.

Conclusions

The hypothesis presented here, that the organic matter in carbonaceous chondrites formed in the solar nebula under conditions of thermodynamic equilibrium, certainly accounts for most of the organic compounds identified thus far. Further calculations and experiments will show whether it can account for *all* the organic compounds in meteorites, or whether additional nonequilibrium or biological processes must be postulated.

Obviously, this hypothesis may also account for the organic matter on comets and inner planets. The latter possibility is particularly intriguing. Perhaps a significant part of the prebiological organic matter on the earth was made, not by Miller-Urey reactions in the earth's atmosphere, but by equilibrium processes in the solar nebula (*27*). If the earth began its history with a substantial endowment of complex organic matter, the chemical evolution stage leading to the origin of life would have had a long head start.

The carbonaceous chondrites Orgueil, Murray, and Cold Bokkeveld contain H_2, CH_4, CO, CO_2, NO, N_2, SO_2, and CS_2; benzene, toluene, naphthalene, anthracene, and other aromatics; sulfonic acid esters; and chlorinated hydrocarbons. Ethane and its homologs are present at $\leq 10^{-3}$ the abundance of methane. Ammonia is likewise undetectable. Neither Miller-Urey reactions nor biological processes seem capable of accounting for this distribution, particularly for the high methane-ethane ratio and the preponderance of aromatic hydrocarbons. However, the observed distribution agrees remarkably well with a distribution calculated by Dayhoff and her co-workers for conditions of thermodynamic equilibrium in a mixture of carbon, hydrogen, oxygen, and nitrogen at 500°K. Apparently the organic compounds in carbonaceous chondrites were formed by equilibrium processes in the solar nebula, after most of the hydrogen had been lost. Some of the prebiological organic matter on the earth may have originated in the same manner.

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