um and then "soaked" by exposure to air for times up to several months. Our initial experiments were done by "soaking" air-equilibrated amber in pure Ar at atmospheric pressure. We found that soaking particles of 1.3-mm mean diameter for 6 days reduced the N_2/Ar ratio from the initial value of 39 to 1.30. Further, soaking our smallest grains (less than 0.18 mm) in Ar for 4 days reduced the ratio from 39 to a value as low as 0.45. Soaking the amber to solubility equilibrium, followed by vacuum extraction in successive stages, provided accurate measures of the diffusivities of the gases. Thus amber fragments soaked in air for several months after crushing were used for diffusion measurements by sealing the fragments in glass tubes after rapidly pumping off the air, and then reopening the tubes periodically with a flexible vacuum tube-breaker, collecting the exsolved gases, and resealing the tube.

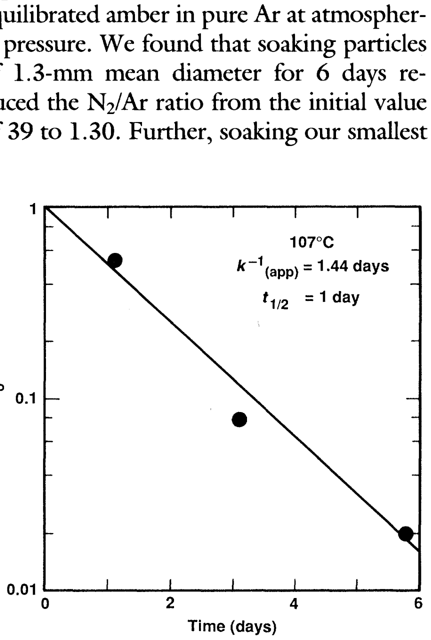

Fig. 1. O_2 consumption by freshly ground Baltic amber at 107°C in a closed system with air. The ordinate shows the concentration of O_2 in the tube versus that at the start, plotted versus time. The consumption rate is first order with a reaction (plus diffusion) "apparent" half-time of 1 day. The "apparent" rate constant $k = k^{*} \cdot A \cdot M$, where A is the specific area (equal to $6/\rho d$ for spheres, where ρ is the density and d is the mean diameter), and M is the mass of amber used. In these experiments d is estimated to have been ~50 μm, and M was 0.10 g; thus $A \cdot M \approx 116$ cm^2 and the actual rate constant $k^{*} \approx 1/167$ days cm^2.

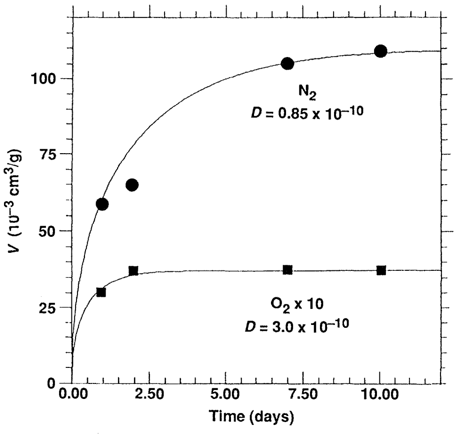

Fig. 2. Diffusion coefficients, D, in Baltic amber at room temperature. Data points are the volumes of gas (V) diffusing out of air-equilibrated crushed amber (40-80 mesh, mean diameter 0.276 mm) into an evacuated sealed glass tube, plotted as cumulative amounts versus time. The curves are calculated (*6*) for spherical fragments exsolving gases to vacuum. The total O_2 content [3.75×10^{-3} cc(STP)/g] is only 8.5% of that expected from the N_2 and Ar values (see text), indicating removal of O_2 within the amber by oxidation reactions. Argon data (not shown) gave a value of D equal to that for N_2: note that $D(O_2)$ is ~ 3.5 times larger than $D(N_2)$ in amber.

Figure 2 shows the results of one set of measurements on N_2 and O_2 in Baltic amber fragments over 10 days. The curves are calculated for spheres of the mean diameter exsolving gas by diffusion into vacuum (*6*). These fragments are heterogeneous media, as indicated by the total N_2 concentration (110 mcc/g), approximately twice the atmospheric-equilibrium solubility, which, from the correlation with the measured N_2/Ar ratio of 52.4, corresponds to half the total N_2 being present in bubbles. Note that the O_2 content, 3.75 mcc/g, is much less than the concentration expected from the N_2 and Ar contents, 44 mcc/g. That is, even after soaking in air for several months, *less than 10%* of the O_2 that has diffused into the amber can be re-extracted by diffusion. We interpret this as indicating that O_2 is actively consumed *within* the amber grains as well as on the surfaces.

Even more significantly, the experimental results in Fig. 2 show that the diffusion coefficients, on the order of $D \sim 10^{-10}$ cm^2/s, are much too high for the gases in amber to be antiques. From the diffusion equation for spheres (*6*) we define an *e*-folding "mean-diffusion time" (that is, the time for reducing the gas contents of the particles to 1/*e* of the initial values) $\tau = d^2/71.7D$, where d is the mean diameter. Thus τ is an effective time constant for considering the diffusive interchange of gases between spherical lumps of amber and the surrounding medium. For times on the order of 50×10^6 years, we require $\tau \sim 2 \times 10^8$ years in order to preserve most of the gas. For a 10-cm diameter piece of amber, we require D as low as 2×10^{-16}, about six orders of magnitude smaller that the value found for O_2 (Fig. 2, lower curve). In this case we require a ball of amber about 116 m in diameter to preserve the initial gases!

This calculation is extreme, of course, because we assumed zero gas pressures in the external medium, although this is nearly the case for amber buried in seawater for most of its existence. Nevertheless, these results show that the air in amber is very young. While the diffusion coefficients are not yet completely understood with respect to the heterogeneous nature of the amber (*7*), the orders of magnitude are clear. With D for air ~10^{-10}, the air inside a 10-cm ball of amber is about 440 years old; in a 1-cm ball it is only about 4 years old. Regardless of the stability of O_2 in bubbles, gases in amber cannot tell us anything about air older than a few centuries. If amber is "forever" (*8*), its occluded air supply, unfortunately, is not.

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3. The atmospheric N_2/Ar ratio is 83.6: the ratio at atmospheric solubility equilibrium is 38 in water and, as we determined, ~39 in amber.
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5. Berner and Landis identified gases in two phases in amber, but they assumed that gases in these phases (matrix and bubbles) are completely independent entities. However, our data on diffusion rates show that gases in bubbles will be in equilibrium with gases dissolved in the matrix, barring very rapid chemical reactions. Their finding that Dominican amber bubbles contained gases at pressures of 10 atm was based on total extraction of gases from pieces in which large bubbles of measured volume were observed. However, the N_2/Ar ratios show that only about half the gas is in bubbles, and most of the gas must be in bubbles too small to measure as well as dissolved in the matrix. Since they did not report gas concentrations, we cannot explain their results in detail.
6. H. S. Carslaw and J. C. Jaeger, *Conduction of Heat in Solids* (Oxford Univ. Press, London, ed. 2, 1959), case IV, p. 235.
7. For example, measurements on another piece of Baltic amber in which the N_2/Ar ratio showed that no bubbles were present gave $D(N_2) = 2 \times 10^{-10}$, whereas we would expect that with no bubbles D would be less than the value for the bubble-rich sample in Fig. 2.
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Response: Since publication of our report (*1*), we have conducted more than 300 gas analyses on 15 ambers and resins that range in age from lower Cretaceous to recent time and are geographically distributed from many of the known world occurrences. We find that by our successive crush technique (*1*) we can usually see mixing trends between two or more compositionally different gases in amber, with one end member containing high values of O_2 and N_2, and the other end members being generally anoxic and enriched in CH_4, CO_2, H_2, and organic gases. In samples where Ar can be discriminated from organic gases, the O_2-rich end member is found to exhibit airlike N_2/Ar ratios. From detailed petrographic study and published analyses of gases within modern tree trunks (*2*), we hypothesize that the non-air gases represent "tree gas" dissolved in amber and derived from the original tree (N_2 + CO_2 + Ar + minor O_2 in a few samples) and "amber gas" formed during the maturation of the amber (CH_4 + CO_2 + H_2 + organic gases). Two examples of gas mixing trends are shown in Fig. 1. Figure 1A shows the existence of two mixing trends within the same sample, which argues persuasively against diffusional exchange with the modern atmosphere.

We also have found that the maximum measured or extrapolated end-member values for O_2 concentrations in Cretaceous ambers are consistently greater (averaging 35% O_2 on an O_2 + N_2 basis) than for Tertiary ambers and modern resin, which give extrapolated or maximum measured O_2 values similar to those in the modern atmosphere. We have measured actual O_2 concentrations greater than 25% in more than 20 individual crushes of five different Cretaceous ambers. We generally find large crush-to-crush compositional variations (Fig. 2) that we attribute either to mixing with the anoxic gases or to partial reaction of O_2 with the original resin. We can now show, on the basis of gas mixing trends, that our low O_2 results reported for Dominican amber (*1*) are probably due to a reaction of O_2 with resin. Also, we now believe that CO_2 derived from original respiration within bubbles (*1*) is of secondary importance compared with CO_2 derived from tree gas or decarboxylation accompanying resin maturation (*3*).

Our analytical technique (*1*) involves gentle breakage of each piece of amber into a few fragments, resulting in the release of small amounts (10^{-10} to 10^{-12} mole) of gas. Successive turnings, in small increments, of the crank attached to the crushing piston, break each fragment into a few smaller fragments. This method of gas release, combined with petrographic and scanning electron microscope studies of the gas bubbles in amber, permits a site-specific analysis of gas in amber not possible with bulk crushing or with grinding methods such as those used by Horibe and Craig (*4*). When present in the amber, gas of larger bubbles (tens to hundreds of microns in diameter) is analyzed during the first few crushes. Gases released by breakage are removed from the crushing chamber on a time scale of tens of milliseconds (not seconds, as assumed by Hopfenberg *et al.*) and recorded by the quadrupole mass spectrometer in real time.

The peak intensity at each mass is the net difference between a maximum just after the gas release and the immediately preceding background. This method allows us to distinguish gas immediately released to vacuum by the opening of bubbles and the generation of new internal surfaces from gas continually desorbing from the exposed surface of the amber and the walls of the vacuum system. Also it minimizes gas loss by back-adsorption on the amber or on the walls of the enclosing vessel. Because of the small quantity of gas released with each crush, complete spectra (average of 30 measurements on each peak) are taken within 40 ms and repeated every 180 ms to capture the gas-release profile. Less rapid gas sampling and vacuum handling procedures are fraught with many problems resulting from back-adsorption during quantitative analysis (*5*). This rapid and precise method allows us to discriminate between very small amounts of O_2-enriched gas released during initial crushing ("air") and the other anoxic gases which are orders of magnitude more abundant and which dominate later crushing. Also, we use a tandem quadrupole geometry with an intervening liquid nitrogen cryotrap baffle placed between the two mass spectrometers. Normally, gases are analyzed only by the first spectrometer. However, occasionally we checked the mass spectra emerging from the second spectrometer and, because of removal of possibly interfering organic gases in the cryotrap, we were able to verify that our gases were indeed rich in O_2.

We have performed two separate experiments similar to those of Horibe and Craig and of Hopfenberg *et al.*; however, our kinetics of gas release cannot be explained simply as diffusion according to the appropriate expressions of Carslaw and Jaeger (*6*). In our experiments, gas release with time was monitored on the quadrupole mass spectrometer and with a capacitance manometer. We used 8 to 20 Cedar Lake amber grains of nominal 2.0-mm diameter that had not been cryoground or otherwise mechanically reduced. In the first or static experiment, amber grains were pumped for 2 hours, attaining 1.3×10^{-8} torr vacuum before the 10-cm^3 chamber was isolated; the rise in pressure was monitored continuously for 5.5 days with a capacitance manometer. In a second experiment, gases released from Cedar Lake amber with time were continuously pumped, kept at about 10^{-8} torr, and recorded by the quadrupole mass spectrometer so that we could obtain continuous compositional information. After the first day, the composition of desorbed gas was virtually constant and identical to that of background gas released from an empty chamber. Gas release followed kinetics of desorption (linear with time) and not the $t^{1/2}$ dependence expected for diffusion (*7*). If our data are interpreted as diffusion, the two experiments give diffusion coefficients different by a factor of 10^5. In sum, we feel our results are best explained by adsorption-desorption phenomena and not by volume diffusion.

We conclude that adsorption and desorption of gases, from both amber samples and from the walls of the enclosing vacuum system, preclude unambiguous interpretation of gas uptake and release experiments to measure diffusion rates in amber, both in our work and in that of our critics (*8*). Uptake and release of gas from even the walls of empty vacuum chambers are slow and can continue for days (*9*). The mechanical grinding of amber, as used by Horibe and Craig and by Hopfenberg *et al.*, further complicates matters by producing surface disruption and a variety of mechano-chemical phenomena that greatly enhance surface sorption of gases (*10*). Even cryoground samples are not free of such effects. Although the half-life of free radicals created by disruption of bonds (*11*) is not great after crushing or grinding, or both, the substance often is permanently modified at the surface, with increased surface area, greater reactivity, increased capillary condensation of gas-

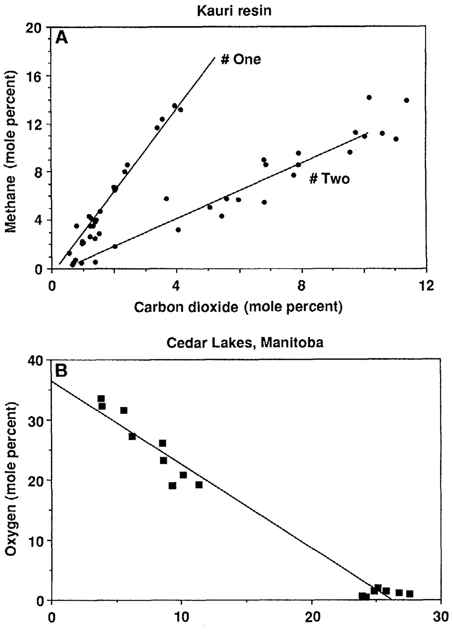

Fig. 1. (**A**) Gas composition data from modern kauri resin. Two separate trends illustrate mixing of methane- and carbon dioxide–free air gases with anoxic amber curing gases and "tree gases." (**B**) Cretaceous Cedar Lakes amber gas data illustrating a mixing line with oxygen and carbon dioxide. Atmospheric gas should be essentially free of CO_2, whereas "tree gas" is CO_2-rich. The intercept from the least-squares regression (36.5 mole percent O_2, $r^2 = 0.98$) indicates the O_2 level of the atmospheric end member.

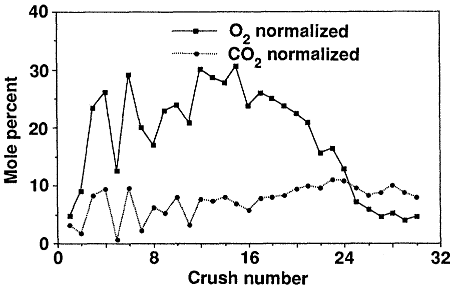

Fig. 2. Raritan, New Jersey, amber gas data illustrating crush-to-crush variability. Oxygen and carbon dioxide mole percent, normalized to the sum $N_2 + O_2 + CO_2$, are plotted as successive crushes of the same sample. We interpret this variation as resulting from mixing between air-derived and anoxic gases, or partial reaction of the oxygen with the original resin, or both.

es, and enhanced physical and chemisorption (*12*).

Surface concentrations of a diffusant in contact with its vapor (adsorption amount) must be known before an experiment can yield a diffusion constant (*13*). For example, in the experiment of Hopfenberg *et al.*, ambiguities arise in interpreting their results. Their choice of propane is understandable when one considers that gravimetric measurements were their basis of monitoring gas uptake. However, as with any common organic gas below its critical temperature, propane will readily condense as multilayer coverage (*12*, *13*). Further, on nonpolar amber surfaces, such organics readily form hemimicelle coatings many multilayers deep (*14*). This results from London–van der Waals surface forces combined with the interaction of adjacent chains of CH_2 groups in organic molecules. Perhaps for this reason their amount of gas uptake was at least 400 times that of our similar release experiments in which we monitored desorption of air gases. Amber resins are polymerized within months of exposure (*15*) and develop detectable polymer cross-linking bonds or side packing of polymer chains, as indicated by broad x-ray diffraction at about 6.3 angstroms and by the anisotropic optical properties of many ambers observed in polished section. A Dominican amber we attempted to chemically analyze proved to be only about 40% soluble in organic solvents. These facts indicate long-range structural order of the amber polymer, which should inhibit rapid diffusion. Finally, Hopfenberg *et al.* suggest that since propane uptake experiments with both ground amber and microtomed amber of largely differing area/volume ratios yielded similar diffusion coefficients, the process involved must be one of absorption and not surface adsorption. In fact, the area to volume ratio in both experiments is nearly the same (*16*).

Regarding the comments of Beck, (i) heating of cloudy amber in oil undoubtedly is done to reduce its viscosity at temperatures above that of boiling water and to allow simple gravitational evasion of bubbles (*17*). This says nothing about diffusion at room temperature. (ii) Excess pressures of gas inclusions within minerals from buried sediments are common (*18*) and, because burial pressures are lithostatic (analogous to burial in a column of water), there is no reason to expect bubble flattening. In our petrographic studies we found that ovoid bubbles in amber often paralleled flow lines and are likely to be the result of deformation during flow, with setting of the amber before the bubbles can regain their spherical shape. The anisotropic optical extinction of amber circumferential to bubbles was observed under the microscope with crossed nicols and indicates strain in the amber due to greater than atmospheric internal bubble pressure and the drop of homogeneous lithostatic burial pressures. In fact, many bubbles in amber have less than atmospheric pressure, as the gases have reacted with the amber after entrapment (*18*, p. 325). (iii) The amounts of CH_4, CO_2, H_2, and organic gases that we obtained do not correlate with the degree of crushing or amount of new surface area exposed. Higher concentrations of CO_2 and CH_4 were often found in initial crushes where breakage was into only a few fragments, exposed internal surface area was minimal, and larger bubbles were intersected. Furthermore, these gases are also given off by unground amber subjected to vacuum extraction as found in our "diffusion" experiments. Thus, Beck's contention that these gases are entirely produced by crushing does not agree with our observations. Much of the gas must be in the amber matrix or in bubbles.

Even if our own experiments and those of Horibe and Craig and of Hopfenberg *et al.* actually demonstrate rapid bulk diffusion, there are still a number of unexplained observations in our work. (i) We found a nonhomogeneous gas distribution within each of our amber samples which could not exist if diffusion were rapid. In other words, there were point-to-point (crush-to-crush) variations in chemical composition and total mass of gas (Fig. 2), and these cannot be explained by an equilibrium solubility distribution or by production of gases by crushing. These data are better explained by mixing of different gas types in the amber. Also, high O_2 concentrations were found only in early crushes, which does not agree with simple O_2 solubility in the amber matrix as discussed by Hopfenberg *et al.* (ii) We found appreciable concentrations of gases such as CO_2, CH_4, and H_2, which should have been lost to the surrounding air by diffusion. (iii) No correlation exists between amber gas composition and time of exposure to modern air. In our study, exposure times ranged from a few years for a drill hole sample to probably hundreds of years for material found on beaches. (iv) We found no O_2 in modern kauri resin that exhibited no bubbles, whereas in the same resin with bubbles we found variable concentrations ranging up to 21%. (v) We found no appreciable difference in O_2 content of gases from the outer reddened and cracked portion of a sample of Baltic amber and the interior relatively clear uncracked portion. This result indicates that reddening of amber does not involve alteration of the gaseous composition. (vi) Most important, there was a consistent relation between O_2 content and geologic age for 15 different ambers from all over the world, with the Cretaceous ambers exhibiting higher O_2 contents than the Tertiary ambers and modern resins.

We admit that our diffusional critics have made an important point, and we thank them for this. Nevertheless, we would be more convinced if further diffusion measurements, free of possible complications resulting from grinding or from adsorption-desorption, were made. An experiment that would better determine the diffusion coefficients for "air" gases in amber would be the demonstration of the penetration of radiolabeled or isotopically defined gas into bulk amber (not along cracks) that would readily be interpretable by its spatial distribution as diffusion. If diffusion is important, we can construct a possible senario to explain some of our observations. If diffusion is moderately fast, but slower than that reported by our critics, it is possible that modern air diffuses into more permeable sites that are more easily accessible by initial crushing. Also, older ambers may have greater O_2 absorptivity than younger ambers, or they may undergo similar diffusion but slower loss of O_2 by reaction with the amber. We see evidence for partial reaction of O_2 with amber in several of our samples, both ancient and modern. Our original idea was that O_2 reaction may occur during the early resin stage of amber formation, but that further reaction is inhibited by slow diffusion within the amber. If diffusion is as fast as our critics say, we must abandon this hypothesis. Intuitively one would not expect gases in amber bubbles to be preserved for millions of years, but our measurements so far have gotten in the way of our intuition.

It is important to point out a misconception that might arise if the gases in amber are indeed not preserved from ancient times. It is that there still exists a substantial amount of independent evidence that O_2 probably has changed over Phanerozoic time. The residence time for O_2 in the atmosphere is only about 7 million years, and the major processes of production (burial of organic matter and pyrite) and consumption (weathering of organic matter and pyrite) have undoubtedly changed in rate over geologic time. This conclusion is based both on the carbon and sulfur isotopic composition of sedimentary rocks (*19*) and on the chemical composition (*20*) and distribution of different sediment types (*21*) over time. For example, recent calculations (*21*) show that redistribution of major worldwide sedimentation, from coal basins to red beds, at the end of the Permian Period must have caused an appreciable lowering of atmospheric O_2 at that time. We believe the atmosphere has evolved along with life and has exerted an influence on life as well as on surficial processes such as forest fires. In this way we are in agreement with the general thrust of the comments of Chave and Smith (although we disagree on details). On the other hand, we disagree with the statement by Hopfenberg *et al.* that "previously held views of paleontology, geology, and atmospheric science" are still intact. These "previously held views" are now undergoing revision independent of our amber studies!

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