through rock-water interaction in the sediment experiment had ceased by 768 hours and the component had been redistributed between the zones. The large apparent Soret coefficients of magnesium and calcium from the sediment experiment relative to the pure values from the control experiment may be attributed to continuous depletion of these elements in the hot zone of the sediment experiment. This is consistent with observed formation of clay and anhydrite alteration phases.

Results of the hydrothermal experiments show that the fluid chemistry in the vicinity of a heat source in pelagic clay or other geological media of low permeability will be influenced by a combination of rock-water interaction and diffusional effects. In particular, a thermal gradient in this environment indicates thermal diffusion which results in large-scale and relatively rapid fluxes of aqueous components from the hot zone toward the cool zone. This "unmixing" serves to decrease the ionic strength in the hot zone by removing a large proportion of sodium and chlorine and may inhibit development of acidity in the near-field region through the removal of silica. The effect on rates of canister corrosion and radionuclide transport requires further evaluation. Magnesium, calcium, potassium, and sulfate, as well as a variety of minor elements, show a strong tendency to participate in temperature-dependent rock-water interaction processes as well as thermal diffusion. The extent to which this occurs depends greatly on the initial rock or sediment composition, solution chemistry, and time. Assessment of these processes is critical for modeling of the near-field chemical environment and mass transport from the near to the far field. The role of thermal diffusion, in particular, must be thoroughly evaluated in any program involving the disposal of high-level radioactive waste in geological media of low permeability.

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The Tunguska Explosion of 1908: Discovery of Meteoritic Debris near the Explosion Site and at the South Pole

Abstract. *Submillimeter-sized metallic spheres extracted from soil in the Tunguska region of central Siberia contain noble metals in cosmic proportions. The trace element composition and geographical distribution of these spheres suggest that they are from the 30 June 1908 Tunguska explosion and not meteoritic ablation products falling continuously on the earth. Debris from this explosion was also discovered in a South Pole ice core; this discovery indicates that the Tunguska object exploded in the atmosphere with subsequent stratospheric injection and transport of the debris. The celestial body that exploded over Tunguska weighed more than 7 million tons, was more than 0.16 kilometer in diameter, and may well have been a stony meteorite. This discovery offers a new precision time marker in polar ice strata for the year 1909. The steady-state influx of cosmic matter at the South Pole is estimated to be* 1.8×10^{-8} *grams per square centimeter per year, which corresponds to a global influx of* 4×10^5 *tons per year.*

On 30 June 1908 at 7:17 a.m. local time (12:17 U.T.), a great explosion occurred in the sky over the basin of River Podkamennaya Tunguska in central Siberia (60°55′N, 101°57′E). Since then, many expeditions have been undertaken to the site and many books and reports published (*1–5*). No crater has ever been discovered during any of these expeditions, a mystery that has generated considerable speculation. The explosion was enormous. It was observed over a radius of 600 to 1000 km in central Siberia, sunlight reflected from the debris lit up the night sky for several days over Europe and western Asia, and trees were blown down over an area of several hundred square kilometers near the epicenter. Because of the lack of obvious clues expected from the impact of a celestial body (an impact crater or identifiable meteorite fragments), various explanations for the origin of the explosion have been proposed.

Careful, painstaking fieldwork by Soviet scientists eventually uncovered some important clues. Soil samples from several areas were subjected to density and magnetic separations and were found to contain black and shiny microscopic metallic spheres (*1, 2*). When scientists examined the geographical distribution of the spheres, they noticed an increasing concentration of spheres northwest of the epicenter (1, 14, and 33 spheres per unit area at 25, 40, and 70 km, respectively), whereas the concentration in the southeast direction was 7, 3, and 10 per unit area (*3*). They concluded that the Tunguska object exploded in midair and that the metallic spheres were carried by the prevailing wind, producing the observed distribution. [The wind direction of the day was from southeast to northwest (*1, 2*)].

I have examined eight of these spheres by high-sensitivity neutron activation analysis with an intent to answer the following questions: Are they truly extraterrestrial? Are all the spheres related to each other as would be expected if they originated from the 1908 explosion? Can these spheres be distinguished from the normal cosmic ablation products of iron meteoroids? (*6*). The abundances of all the elements measured in these spheres are shown in Table 1. Iridium, a reliable indicator of extraterrestrial matter, is unmistakably enriched in all eight spheres. In addition, they all contain nickel and cobalt, two other "metal-loving" elements always found with iridium in cosmic matter. Five of the eight spheres have lower and similar concentrations of iridium, nickel, and cobalt; three are characterized by very high concentrations of these elements. The iridium content of sphere H is 56,900 parts per billion (ppb) [only one iron meteorite is known with a comparable iridium content (Negrillos meteorite with 45,000 ppb iridium and 5.32 percent nickel (*7*))]. The average Ni/Ir ratio (by weight) in the first six spheres listed in Table 1 is identical to the cosmic ratio, 2.0×10^4 (Fig. 1). For the last two spheres the Ni/Ir ratio is

Table 1. Abundance of elements in eight metallic spheres recovered from the region where the Tunguska object exploded in 1908. Uncertainties in the abundances represent 2σ counting statistics (σ is the standard deviation). Abbreviations: ppb, parts per billion; ppm, parts per million.

Sample	Weight (μg)	Nickel (%)	Cobalt (ppm)	Iridium (ppb)	Gold (ppb)	Chromium (ppm)	Antimony (ppb)	Iron (%)
A*	43	0.062 ± 0.014	153 ± 5	25 ± 8	11 ± 4	76 ± 2	1,900 ± 200	78
C	69	0.170 ± 0.019	676 ± 20	69 ± 10	8 ± 15	5 ± 2		81
D	60	0.063 ± 0.015	107 ± 3	55 ± 8	≤5	8 ± 2	210 ± 60	76
E	74	0.036 ± 0.009	115 ± 3	29 ± 5	17 ± 5	9 ± 1	200 ± 70	76
G	36	0.093 ± 0.013	233 ± 7	45 ± 7	7 ± 6	11 ± 1	640 ± 120	78
F†	59	6.07 ± 0.08	2,580 ± 80	2,500 ± 40	216 ± 24	3,770 ± 15	≤130	78
B‡	111	4.63 ± 0.10	4,280 ± 130	9,040 ± 70		393 ± 12		77
H§	55	21.4 ± 0.2	11,900 ± 400	56,900 ± 220	420 ± 80		≤290	81

*Contained 4.4 ± 0.7 ppm arsenic. †Contained 53 ± 55 ppb rhenium and 3.8 ± 1.2 ppm osmium. ‡Contained 430 ± 280 ppb rhenium, 34 ± 4 ppm osmium, and 40 ppm platinum. §Contained 4,300 ± 600 ppb rhenium, 83 ± 5 ppm osmium, and 80 ppm platinum.

less than the cosmic value and may result from partial evaporation or, more probably, incomplete condensation of nickel, since nickel is more volatile than iridium.

The gold content of the spheres correlates with the nickel content: low in the nickel-poor spheres and high in the nickel-rich spheres. The trend in the antimony concentration, however, is just the opposite. The Sb/Ir ratio in the low-nickel spheres varies from 4 to 76 compared to the cosmic Sb/Ir ratio of 0.27; evidently the spheres are enriched in antimony. The iron content ranges from 76 to 81 percent. For sample H the finding of 81 percent iron and 21 percent nickel implies that these elements are largely in the metallic state with only a trace of Fe_3O_4 on the surface of the sphere (observed in my scanning electron microscope study). Sample H also contains other refractory noble metals such as osmium, rhenium, and platinum in cosmic proportions.

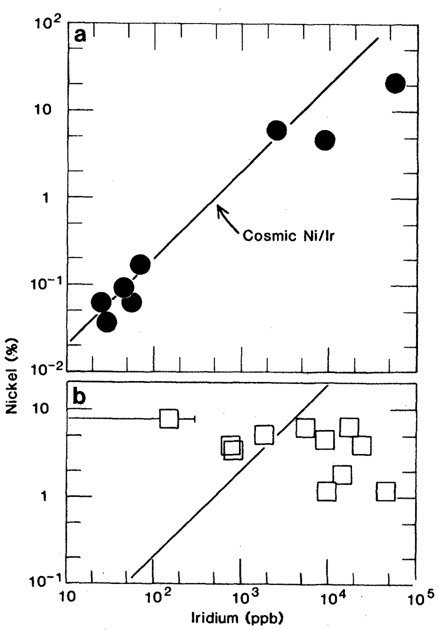


Fig. 1. (a) Iridium and nickel, the two diagnostic indicators of extraterrestrial matter, not only are well correlated in the Tunguska spheres but are present in cosmic proportions, an indication that all these spheres originated from the same celestial body. (b) Metallic spheres from daily, meteoric ablation, in contrast, do not show any such correlation, mainly because they are products from compositionally different meteors over many thousands of years.

These data establish that all eight spheres are extraterrestrial. Let us now examine whether they originated from the 1908 explosion or are derived from the steady-state rain of meteoritic ablation spheres. Figure 1 compares the distribution of nickel and iridium in the Tunguska spheres with that of ablation spheres separated from 2 kg of red clay sediment from the mid-Pacific Ocean. There is no correlation between nickel and iridium in the 11 black and shiny metallic ablation spheres. Although the iridium content in these ablation spheres varies by a factor of 1000, the nickel content varies by less than a factor of 10.

The lack of correlation between nickel and iridium in the ablation spheres may be due to two factors: (i) these spheres come from compositionally different iron meteors whose iridium concentration varies much more than their nickel concentration and this collection represents all the products over thousands of years and (ii) chemical processes associated with the melting of ablation products could vary the relative proportion of these two elements. In contrast, the correlation between nickel and iridium in the Tunguska spheres and their cosmic Ni/Ir ratio indicate that these spheres came from a single object that contained the nonvolatile elements in cosmic proportions. For cosmic ablation spheres found in sediments, there is a hint that these are residues from the evaporation of volatiles. Spheres rich in refractory iridium are usually low in nickel.

The trace element data also provide information on the conditions under which sphere formation took place. The abundance pattern of trace elements relative to carbonaceous chondrites in spheres A and H, characteristic of low- and high-nickel spheres, are shown in Fig. 2. Sphere H is enriched in refractory elements (rhenium, osmium, iridium, platinum), whereas sphere A is depleted in refractory elements but enriched in the more volatile elements (arsenic and antimony). The most straightforward explanation is that sphere H acquired its full complement of refractory metals during condensation but ceased to equilibrate with the impact vapor cloud before much of the cobalt, nickel, iron, and all the more volatile elements condensed. By contrast, sphere A acquired its full complement of all elements less volatile than gold plus a substantial amount of nonmetallic material (presumably iron oxide). These results suggest that, in the region of space where the Tunguska object exploded, the temperature was high enough to melt and vaporize the entire object.

It has been estimated that the mass of

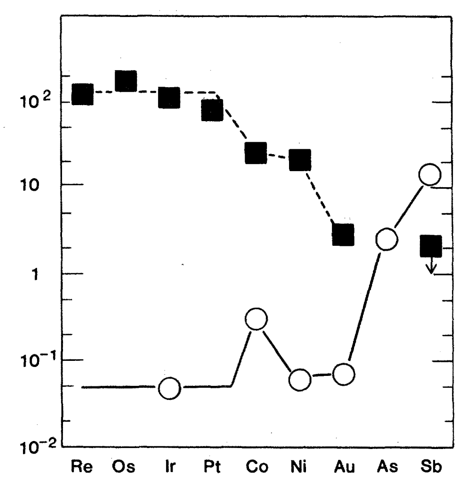
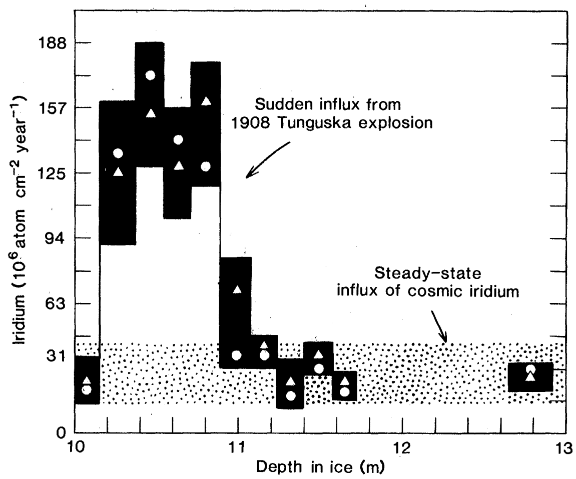


Fig. 2. Trace element abundances relative to carbonaceous chrondrites in the low- and high-content nickel spheres (spheres A and H, respectively) from the Tunguska region. The observed abundance patterns reflect condensation of these spheres at different temperatures. The patterns for sphere H represent material condensing at very high temperatures when volatile elements such as arsenic and antimony are still in the vapor phase.

Fig. 3. Abundance of iridium in the 101-m Antarctic ice core at the South Pole. The iridium contents calculated with the use of the 317- and 468-keV gamma rays from ^{192}Ir are plotted separately (triangles and circles). The shaded band represents iridium from the steady-state influx of cosmic matter, 29×10^6 atom cm^{-2} $year^{-1}$. Excess iridium from the 1908 explosion is present in the ice core at a depth of 10.15 to 11.07 m. The layer from 10.89 to 11.07 corresponds to the year 1912 ± 4 years on the basis of microparticle dating. The discovery of iridium from the Tunguska event in this layer suggests that the actual age is 1908–1909. The excess iridium over the steady-state background corresponds to 3.5 tons of iridium distributed globally.



the Tunguska object was about 10^6 tons (*3*). Its failure to produce a crater, coupled with its cosmic composition, the presence of metallic and glass spheres fused together (*1*), and the high abundance of chromium in some metallic spheres argue against an iron meteorite projectile.

The eyewitness accounts of the Tunguska explosion (*1, 4*) together with my findings demonstrate that the Tunguska object was vaporized by the explosion in the atmosphere. The question of whether the debris from this event could have reached the stratosphere and as a consequence been distributed globally prompted me to search the debris from this event in the Antarctic ice. It is well known that fission products from nuclear explosions carried out at ground level reach the stratosphere (*8*). We do not know the energy and force characteristics of the Tunguska explosion, nor do we know how similar it was to a nuclear detonation. However, the discovery of Tunguska debris in Antarctic ice would not only confirm injection into the stratosphere and subsequent transport but would also provide independent evidence for an explosion of a cosmic body in the atmosphere in 1908.

The average accumulation of ice (in water equivalent) at the South Pole is ~7 cm $year^{-1}$ (*9*). Although this accumulation rate in conjunction with the microparticle concentration is a reasonably good chronometer for measuring time with depth, an error of some 90 years in a 911-year record has been estimated (*9*). I selected the 101-m ice core drilled in 1974 at the Amundsen-Scott station because Thompson and Mosley-Thompson (*9*) have studied this core extensively, using microparticle techniques (*10*). Iridium, an ideal tracer with which to detect the cosmic debris from the 1908 explosion, was radiochemically analyzed in all the samples (Fig. 3). The iridium contents were calculated with the use of the 317- and 468-keV gamma rays from ^{192}Ir. Since the iridium concentrations for experimental blanks were quite low relative to the samples in both irradiations, no corrections have been made to the sample data (*11*). The iridium in the insoluble matter separated from Antarctic ice at this depth interval can arise from two sources: (i) steady-state influx of cosmic matter from the interplanetary dust cloud as micrometer-sized particulates or (ii) debris from the Tunguska event. Reading from right to left in Fig. 3, one finds that the first five samples of ice layers, each representing 1 year's accumulation, have an iridium content of 29×10^6 atom cm^{-2} $year^{-1}$. The ice layer on the extreme left also has a comparable iridium content, 24×10^6 atom cm^{-2} $year^{-1}$. Since the extreme layers, corresponding to the years 1920 ± 4 and 1900 ± 4 based on the microparticle dating, are outside the time period of the Tunguska event, it is assumed that they measure the steady-state cosmic influx. The iridium content rises over the steady-state background in the ice layer from 11.07 to 10.15 m. The dates of these layers assigned from microparticle concentration are 1912–1913 ± 4 years and 1918 ± 4 years. The iridium content in this depth interval is greater by about a factor of 4 than the background. The presence of excess iridium precisely in the depth and time interval where Tunguska debris is expected, viewed against the constancy of the steady-state iridium values for the six samples examined here, leads me to suggest that this increase in iridium is due to the debris from the 1908 Tunguska explosion transported here through the stratosphere.

The excess iridium from the Tunguska event at the South Pole is estimated to be 1.6×10^{-13} g cm^{-2}. Because this iridium could only be deposited here by means of stratospheric fallout, it should be present worldwide. The deposition rate at the South Pole is expected to be lower than the global average for two reasons: (i) the South Pole has an unusually low precipitation rate and (ii) the atmospheric circulation pattern preferentially concentrates stratospheric matter in mid-latitudes rather than near the poles. This preferential enrichment factor is somewhere between 3 and 5 (*12*). I have estimated the global fallout of Tunguska debris by multiplying the South Pole data by a factor of 4. This calculation indicates that about 7×10^6 tons of debris fell globally from the stratosphere, if we assume that the Tunguska object is composed of material similar to carbonaceous chondrites. Consequently, the Tunguska object must have been at least 0.16 km in diameter. This estimate is a minimum because the fraction of the Tunguska debris entering the stratosphere is unknown. The fraction of the debris that entered the stratosphere is the most difficult part to estimate because knowledge of the height, explosive force, and meteorological condition are required for this estimate (*13*). Therefore, an upper limit cannot be estimated for the Tunguska body.

It has been suggested that the Tunguska object was a comet or inactive comet (*2, 3*). My estimate of the size of the object is independent of whether it was a common stony meteorite or a comet, since most comets have a ratio of gas to dust of only 1 to 2 (*14*). At present, there are no compelling reasons to believe that the object was a comet. Stony meteorites break up in the atmosphere quite often. Impact craters on the earth less than 1 km in diameter are produced by iron or stony-iron bodies (*15*). The absence of craters in this size range produced by stony meteorites confirms that stony meteorites (<100 m in diameter) are broken up in the atmosphere. A combination of many factors could have contributed to the total breakup of the Tunguska body. But until more is known about the chemical composition of comets, the question of whether the Tunguska object was a meteorite or a comet cannot easily be resolved (*16*).

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10. There are two reliable precision time markers for the years 1884 and 1953 in the firn core. The 26 August 1883 Krakatoa volcanic eruption deposited particulates in the ice core at 14.90 m, and the nuclear explosions of 1953 deposited fission products at 4.35 m. Since these two markers bracket the time period to be studied, the error in dating is now reduced to ±7 years for the 69-year firn core between 14.90 and 4.35 m. Since the time period of my investigation is about 1909 to 1910, the uncertainty in dating a firn layer from this core corresponding to year 1910 is ±4 years. Samples from 10.0 to 11.7 m corresponding approximately to the time from 1920 ±4 to 1908 ±4 years were selected. An additional sample from 12.6 m was also examined. Details on the handling of ice core samples have been described (*9*). One-quarter of the ice core (10 cm in diameter) corresponding to 19.64-cm^2 area was cut into various lengths at the Institute of Polar Studies, Ohio State University. Sodium chloride was then added to the melted ice so as to produce a 2 percent NaCl concentration for conductive purposes. Usually, about ten slices of ice were cut covering a length of 17 cm. After these individual samples were studied for microparticles, the water from them was combined and filtered through special 0.45-μm filter paper. Half of this filter paper was subjected to high-sensitivity neutron activation analysis, and the other half was used to determine the particulate composition in the Polar Research Institute. The filter paper with the particulates was transferred into a specially cleaned high-purity synthetic silica tube, heated slowly to 400°C to destroy the organic matter from the filter, and sealed. These samples were irradiated with standards for 43 days at a thermal neutron flux of 2×10^{14} sec^{-1} cm^{-2}.
11. The procedure for experimental blanks was as follows: 100 ml of deionized water was filtered through a 0.45-μm Millipore filter in my laboratory; this filter was then processed in the same way as the samples. The only difference in the treatment of the blanks and samples was that the blank filter paper was not handled at the Polar Research Institute. The iridium content of the experimental blank was $(1.0 \pm 1.8) \times 10^{-14}$ g.
12. J. L. Barker and E. Anders [*Geochim. Cosmochim. Acta* **32**, 627 (1968)] quoted a factor of 3 for the enrichment at mid-latitudes over the South Pole. L. Machta (personal communication) suggests that it could be a factor of 5.
13. From none to all of the debris from nuclear explosions carried at ground level enters the stratosphere. G. J. Ferber (personal communication) suggests that the percentage entering the stratosphere is highly dependent on the meteorological conditions and explosive yield.
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16. The steady-state influx rate of iridium measured at the South Pole is 9×10^{-15} g cm^{-2} $year^{-1}$ or 4×10^5 tons of cosmic matter per year globally. This is higher by a factor of 4 than the value inferred from mid-Pacific Ocean sediments (*12*). Recent measurements of iridium in deep-sea cores corresponding to the Pliocene and Eocene-Oligocene periods have yielded accretion rates of 3.4×10^5 and 3.3×10^5 tons per year [F. Kyte and J. T. Wasson, *Lunar Planet. Sci.* **13**, 411 (1982)].

17. I am indebted to A. J. Barnard, Jr., J. D. Bonn, T. J. Hurley, H. A. Kaufman, and J. Volkert for support and encouragement; to B. J. Levin, Astronomical Council of the Academy of Sciences of the U.S.S.R., for his assistance in getting the Tunguska spheres; to A. A. Yavnel', Committee on Meteorites, U.S.S.R., for supplying the spheres; to L. Thompson and E. Mosley-Thompson for supplying the particulate matter from the Antarctic ice core and for many stimulating discussions; to A. Meyer and S. Gunn for support in the neutron irradiations; to O. K. Manuel for carrying out the reirradiation of iridium samples after chemical purification; to G. Downing for assistance; and to J. W. Larimer for helpful review.

26 January 1983

Acquisition of Digestive Enzymes by Siricid Woodwasps from Their Fungal Symbiont

Abstract. *Larvae of the woodwasp,* Sirex cyaneus, *contain midgut digestive enzymes that enable them to utilize the major fungal and plant polysaccharides found in their food. At least two classes of enzymes, the* C_x*-cellulases and the xylanases, are not produced by the larvae. Instead, larvae acquire these enzymes while ingesting tissue of* Amylostereum chailletii, *the fungal symbiont that occurs in the wood on which the larvae feed.*

Woodwasps (*1, 2*) maintain a close association with a fungal symbiont both as larvae and adults. Adult female woodwasps oviposit in dying and dead standing trees. As eggs are laid, the wood is simultaneously inoculated with a mass of fungal oidia which are maintained in special pouches associated with the egg-laying apparatus. The fungus permeates the surrounding wood, and the larvae tunnel into this, ingesting both wood and fungal hyphae. Although attempts to study the enzymatic characteristics of the gut fluids of woodwasps have given ambiguous or conflicting results (*3, 4*), Müller demonstrated significant digestion and assimilation of wood constituents, including cellulose and hemicellulose, by larvae of *Sirex gigas* and *S. phantoma* (*4*).

We have found (Table 1) that the midgut of *S. cyaneus* larvae contains enzymes active against a number of plant polysaccharides, including microcrystalline cellulose (the cellulase complex), carboxymethyl cellulose (C_x-cellulase), xylan (xylanase), pectin (pectinase), and amylose (amylase). The midgut of *S. cyaneus* also contains enzymes active toward laminarin, a β-1, 3-glucan representative of a widely distributed class of fungal cell wall polysaccharides. The enzymes are present in larvae that have been reared on balsam fir chips permeated by the mycelium of the fungal symbiont, *Amylostereum chailletii* (*5*), as well as larvae collected from their natural galleries in the trunks of standing balsam fir trees. This same suite of carbohydrases is present in both the culture fluid of *A. chailletii* and in an extract of balsam fir wood permeated by the fungus. These results suggested that *S. cyaneus* might be acquiring essential digestive carbohydrases when it ingests fungal tissue and fungal secretions along with the wood it consumes. In agreement with this idea is the observation that larvae fare poorly when they are fed a diet of symbiont-free balsam fir chips, extracts of which are virtually enzyme-free (Table 1). Larval mortality is high after only a week, and the level of midgut enzymes decreases dramatically (*6*).

In order to test the hypothesis that the enzymatic activity of the larval gut fluid is due to ingested fungal enzymes, we have purified the C_x-cellulases and xylanases from larvae feeding on *A. chailletii*-infested balsam fir chips and from the culture fluid from *A. chailletii* growing on microcrystalline cellulose in a defined medium, and have compared isoelectric points (*p*I) of the insect- and fungus-derived enzymes. C_x-cellulase and xylanase were chosen for detailed comparison since *Sirex* larvae have been shown to assimilate significant portions of cellulose and hemicellulose, which make up nearly 70 percent of the dry matter of balsam fir wood (*4, 7*). Although the presence of enzymes active against pectin, starch, and laminarin suggests that woodwasp larvae have the capacity to utilize polysaccharides other than cellulose and hemicellulose, the limited quantities of starch and pectin in balsam fir wood (*8*) and the sparse growth of *A. chailletii* mycelium in the wood surrounding larval tunnels (*9*) indicate that these are minor sources of nutrients for the larvae.

C_x-Cellulase and xylanase activities were detected in comparable fractions