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## Uranium Series Ages of the Del Mar Man and Sunnvvale Skeletons

Bischoff and Rosenbauer (1) claim that their uranium series ages for the Del Mar man and Sunnyvale skeletons indicate that the previously published aspartic acid racemization ages (2) of these two skeletons are too old. They state that the concordancy of their <sup>230</sup>Th and <sup>231</sup>Pa ages supports the uranium series ages for the bone samples. Uranium series ages, however, are based on the critical assumption that bones incorporate uranium for only a relatively short time after their burial and have subsequently remained closed systems with respect to migration of both uranium and its daughter isotopes <sup>230</sup>Th and <sup>231</sup>Pa. We maintain that the assumption of rapid uranium incorporation followed by closed system behavior is tenuous and that <sup>230</sup>Th-<sup>231</sup>Pa concordancy is a necessary but not sufficient condition to establish this behavior and thus the accuracy of the dates.

Modern bones contain trace amounts of uranium (3). However, fossil bones assimilate uranium during their depositional history (3). [This was one of the three methods (3) used to demonstrate that the famous Piltdown man actually was composed of both modern and fossil components.] A uranium series age is thus the average integrated age of uranium incorporation into a bone; this age is always less than the bone's actual age.

The processes by which bones acquire uranium are complex (4, 5), possibly episodic (6), and poorly understood. As a result, it is not possible to evaluate the rate of uranium accumulation in any particular bone. Bischoff and Rosenbauer assume that uranium is rapidly incorporated into bones by a mechanism that involves the reduction of uranium by "active (or labile) organic matter." No mechanisms have been presented for this process, nor is it known what organic compounds might be involved. The fact SCIENCE, VOL. 217, 20 AUGUST 1982

that fossil bones contain uranium in both the +4 and +6 oxidation states (5) indicates that the mechanism assumed by Bischoff and Rosenbauer does not completely describe how bones assimilate uranium. Moreover, uranium analyses of samples from various stratigraphic units at Olduvai Gorge, Tanzania, have demonstrated that bones and teeth remain an open system with respect to uranium accumulation over periods of hundreds of thousands of years (6, 7).

Bischoff and Rosenbauer state that "it is possible to judge the validity of a uranium series date on a single sample by testing for internal concordancy between two independent decay schemes:  $^{238}U \rightarrow ^{230}Th$  and  $^{235}U \rightarrow ^{231}Pa$ ." In fact, a close examination of the <sup>230</sup>Th-<sup>234</sup>U and <sup>231</sup>Pa-<sup>235</sup>U concordia diagram shows that, for samples with ages of several tens of thousands of years or less, concordancy is a very insensitive test for the validity of the closed-system hypothesis, especially with regard to uranium uptake. For samples at the young end of the concordia curve, late uranium uptake can decrease apparent radiometric ages without disturbing <sup>230</sup>Th-<sup>231</sup>Pa concordancy within experimental uncertainty  $(\pm 2\sigma)$ . For older samples, although concordancy is a better test, what is being dated is the time of uranium uptake by the bone, which is later than their burial time. That the concordancy of <sup>230</sup>Th and <sup>231</sup>Pa ages is not always a valid indicator of whether the resultant ages were equivalent to the burial age of the sample was first discussed by Kaufman (8), who cited concordant ages of 69,000 and 112,000 years that had been obtained for mollusk shells from marine terrace deposits of apparently the same age. Similar problems have been found for bone (9, 10). For example, in the Middle Pleistocene lower stratigraphic units at Caune de l'Arago in the eastern French Pyrenees, concordant (within  $2\sigma$ ) <sup>230</sup>Th and <sup>231</sup>Pa ages range from  $\sim 35,000$  to  $\sim 120,000$  years (10).

As has been pointed out by others (3,9), since the time sequence of uranium accumulation is unknown, even concordant uranium ages are lower limits and should be regarded as minimum ages. Szabo (9) concluded that in some instances concordant uranium series ages for bones can be too young by as much as several tens of thousands of years (11). The uranium series ages given by Bischoff and Rosenbauer for the Del Mar man and Sunnyvale skeletons should therefore be considered minimum estimates, and the actual ages may be greater, possibly by as much as several tens of thousands of years. Such a conclusion is consistent with the racemization ages which have been determined for these skeletons (2).

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- 11. Since submission of this comment we have obtained some preliminary results in a comparative study of uranium series, amino acid racemitwe study of uranium series, amino acid racemi-zation, and collagen-based radiocarbon ages of fossil bones [J. L. Bada and R. Finkel, in *Quaternary Coastlines and Marine Archaeolo-gy: Toward the Prehistory of Land Bridges and Continental Shelves*, P. M. Masters and N. C. Fleming, Eds. (Academic Press, London, 1982)]. Our results confirm Szabo's conclusion; concordant uranium series ages were found to be consistently volumer than the amino acid be consistently younger than the amino acid racemization and radiocarbon ages, in some cases by as much as 10,000 to  $\sim 30,000$  years. We thank H. H. Veeh and our colleagues at Sorine Institution of Oceanoraschi for bollowing 12.
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Bada and Finkel ignore the important fact that our concordant uranium series ages agree with the radiocarbon ages at each of the four sites (1). The best example is the Mountain View dump site, where our age of 19,900 years on a Camelops bone agrees with radiocarbon ages of 20,800 to 23,600 years on seven different samples of wood from the same deposit. The 37,200-year uranium series age on the bone at Rancho La Brea compares well with radiocarbon ages of 35,500 and 36,000 years on collagen from two associated bones. The Sunnyvale burial was intrusive into and, therefore, younger than a bed that yielded radiocarbon ages on two separate shell samples of 10,100 and 10,400 years, consistent with our age of 8,300 years for the skeleton. Radiocarbon ages on shells in the midden from which the Del Mar skeleton was derived range from 4,600 to 12,000 years, consistent with our age of 11,000 years for the skeleton.

Szabo (2) compared uranium series ages on bones with radiocarbon ages of 10,000 to 30,000 years from nine different archeological sites throughout North America. The average difference between the radiocarbon and uranium series ages is 2,700 years. At three of these sites the uranium series ages agreed within counting error with the radiocarbon ages. A large discrepancy was apparent for only one site, where a bone yielded a concordant uranium series age of 18,000 years and a coexisting shell yielded a radiocarbon age of 30,600 years. Even for this sample, the possibility that the single radiocarbon age is in error cannot be excluded. If, as Bada and Finkel maintain, uranium uptake by bone is slow and continuous, such agreement with radiocarbon would not occur.

Bada and Finkel correctly point out that the exact mechanism of uranium uptake is not understood and that bones frequently behave as open systems to uranium. However, their assertion that uranium uptake is always slow and continuous is an outdated theory that in the past led to attempts to use uranium content as a basis for relative dating. The accumulated evidence proves that uranium uptake is not so simple, and relative dating by uranium content has been largely discarded.

For example, the Seitz and Taylor study (3) cited by Bada and Finkel showed that the uranium concentration in bones from Bed II at Olduvai Gorge is significantly higher than that in bones from the underlying Bed I. However, the conclusion of Seitz and Taylor's study, as well as of that by Molleson (4), also cited by Bada and Finkel, was that uranium uptake occurred initially over a short period and that the erratic differences in uranium content with respect to stratigraphic age are due to later and episodic loss of uranium. Such losses give rise to anomalously old ages but also to discordancy between the <sup>230</sup>Th and <sup>231</sup>Pa decay systems.

In giving examples of what they consider to be incorrect concordant uranium series ages on bone, Bada and Finkel change the meaning of concordancy to include samples for which the <sup>230</sup>Th and <sup>231</sup>Pa ages agree within a time span of  $\pm 2\sigma$ . This is twice the range that is conventional among workers in uranium series dating and its effect is to greatly increase the number of apparently concordant ages. As an example, Bada and Finkel refer to concordant ages on bones from the Arago Cave deposits that are too young (5). As part of an international cooperative effort, we analyzed 19 bone samples from the Arago Cave site. Concordancy was claimed for no samples in this study by us or by any of our coworkers. To the contrary, the internal isotopic composition of our samples indicated that gross secondary leaching and migration of uranium series isotopes had taken place in the cave deposits, and we concluded that highly valid ages could not be obtained. We rejected as discordant, for example, a sample that yielded ages of 71,000 years (<sup>230</sup>Th) and 54,000 years (<sup>231</sup>Pa) because these ages disagree by more than  $1\sigma$  (±6,000 years). Bada and Finkel would declare this sample to be concordant because the ages agree within  $\pm 2\sigma$  (12,000 years). Moreover, the excess of <sup>230</sup>Th over <sup>234</sup>U activity in two of our Arago Cave samples from the upper parts of the deposit implies that

the uranium (which is more mobile than thorium) had been lost at a later time and, more important, that uranium series ages can be too old as well as too young. Therefore, Bada and Finkel's conclusion that uranium series ages are always too young is not correct.

We have used concordancy, along with five other internal isotopic parameters (5), as criteria for determining contamination or an open system. Violation of any of these criteria is a basis for rejection of a given age. We do not maintain that concordancy constitutes proof of an age but only that it is a necessary component of our internal criteria for judging an age. In an open system, uranium exchange between bone and surroundings would be erratic over time and, as Veeh (6) has pointed out, uranium addition to or loss from an old sample would cause the <sup>231</sup>Pa and <sup>230</sup>Th systems to move drastically off the concordia plot.

The ultimate test of our uranium series ages involves the geologic and stratigraphic context of the sample and a comparison with other independent radiometric techniques, such as radiocarbon. We consider that we have been consistent and conservative in using these criteria in regard to our dating of the Del Mar and Sunnyvale skeletons and, therefore, we stand by our original age estimates.

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