

in India is not an unusual story. Such things happen all the time in science. For me and many of my colleagues, for instance Pal, Goel and Rama, it demonstrated that we could do good science in India if we had confidence²⁴ in ourselves.

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The discovery of cosmogenic ⁷Be and ¹⁰Be

James R. Arnold

Halfway round the world from Bombay, in Chicago, an independent discovery of cosmogenic beryllium radionuclides.

Willard Libby¹ published his first note on cosmic-ray production of nuclides in the earth's atmosphere in 1946. He followed this in the next few years by developing the ¹⁴C dating method²⁻⁴ and by the measurement of ³H in natural waters⁵, achievements which attracted worldwide attention. Doubtless many persons were led to consider the possibility of finding and using other nuclides made in the same way. Libby himself did so, as he told me years later.

Since N₂ and O₂ are the primary targets in the atmosphere, the list of other interesting products of high yield is short. Among these radionuclides whose half-lives exceed a few hours it has only two entries: ⁷Be (53 days) and ¹⁰Be (1.5 × 10⁶ years). There are many more products from spallation of ⁴⁰Ar but because its abundance is only about 1% they posed a severe challenge to the counting methods available in the early 1950s. Thus it is not surprising that many people were thinking about the Be isotopes, and that some reached the point of doing experiments.

My own interest and taste for such things had of course been aroused by my participation in the ¹⁴C effort. In

addition, I was excited by the possibilities of the then new scintillation-counting technique, and tempted to try my hand at developing a sensitive, low-background system using this method.

In 1952, together with Dr Thomas Sugihara, also a veteran of the Libby group, I had assembled a small NaI (TI) counting system, in a low-background shield, and used it to determine the gamma spectrum and later the decay scheme of natural ¹⁷⁶Lu (ref. 6). This was an interesting radionuclide but there were no immediate applications. The search for ⁷Be seemed more promising, since its half-life and mode of production in the atmosphere suggested usefulness in studying atmospheric circulation and exchange processes. I guessed that it would be found attached as BeO to atmospheric dust, and hence be washed out in rain, much like the bomb fallout that was then so abundant. I did not then know of a number of other efforts, earlier or contemporary, to isolate and detect it. I thought ¹⁰Be even more promising for applications, but more difficult, both because of its long half-life and because it was known to be a pure beta emitter. My plan was to do

⁷Be first.

Rain and snow are frequent in Chicago, so I did not have to go far for samples. I found an old storage building on the university campus with a single drainpipe, and bought a modest supply of tin-plated square five-gallon steel cans. I was also fortunate in hiring a strong and ingenious Iraqi technician, Hussein Ali Al-Salih. When it rained we would sally forth and collect 5-50 gallons of rain water (full of black industrial dust), acidify it, add carrier and lug the cans back to the laboratory. The samples in the cans were well mixed, I'm sure, in transit. Then followed a couple of days of rather messy and sometimes unreliable chemistry, which I devised based on the scattered literature and improved bit by bit. The end result was a small mass of (usually) white BeO powder. The content of ⁷Be in these extracted samples was variable, of course, depending on the details of the history of the air mass and the clouds from which the rain fell. Fortunately in some cases it was quite high, so that extreme low-level methods were not needed, and the identification could be made both from the energy of the



The author counting a ^{10}Be sample using a low-level flow beta-counting system, around 1956

single 478 keV gamma ray and the 53-day half-life. It was gratifying that the amount found was in the right range for cosmic-ray production, though the uncertainties were many.

One incident in this period was memorable. We hoped to be able to obtain samples with less black dust when it snowed, and we could collect the precipitation fresh, without exposing it to a dirty roof. In February 1954 we got our wish, and filled stainless steel buckets all day with snow scooped up as it fell. It sparkled white in the buckets, but turned to the same black colour as soon as we melted it. I hope Chicago is cleaner now.

We wrote the paper up for publication in a happy frame of mind. One point had to be dealt with: could the ^7Be we found actually be bomb fallout, a by-product of nuclear testing, rather than a natural process? The data gave a clear answer, because there was no change after the large H-bomb tests of 1954. But here I ran afoul of security regulations, which required all US scientists to clear statements about nuclear weapons with the Atomic Energy Commission's security office. I could not plead ignorance, since I had been actively involved in the debates that led to the law itself, on which the rules were based. Still I regarded the statement that ^7Be was not made mainly by bombs as not very sensitive, and the approval as a formality.

Just after the paper was complete an important event took place. Prof. S. Chandrasekhar, the famous astrophysicist, invited me to give a seminar on ^7Be to the astronomers at Yerkes Observatory in Lake Geneva, Wisconsin, of

which he was a staff member. I accepted with pleasure. In conversation afterwards, he asked if I had seen the articles on the Be isotopes by Bernard Peters in the *Proceedings of the Indian Academy of Sciences*. I confessed that I did not keep up with this journal, and he gave me a couple of references⁷.

Of course I knew about Prof. Peters' discovery of the heavy ions in the primary cosmic rays, and I knew why he was in India and no longer in the US. I did not then know him personally. The articles in question, with numerous coauthors then unknown to me, were theoretical and analytical, describing the promise of these nuclides, their production rates and inferred distribution in nature, and methods of detection. They made it clear that an experimental effort was in progress.

On my return to Chicago I was disturbed to find a telegram from the security people, directing me not to submit the paper and to keep quiet until they had completed their review—no time-scale indicated. For many reasons this instruction was unwelcome. As I pointed out to them the idea was 'in the air' and (as proved true) others were surely doing similar analyses, the Peters group for one.

I did write and establish contact with Peters, beginning an exchange of ideas and eventually people, which has had benefits for me until this day. Their work was not quite so far advanced, understandably so since nuclear science in India was still very young. Meanwhile I argued with the security people.

Finally an article appeared in which ^7Be and its uses were mentioned without reference⁸. After I called their attention to this, and complained loudly but politely that I was being discriminated against, the security people relented and we were allowed to publish⁹. More active exchanges developed thereafter with the Peters group.

The next step, ^{10}Be , had to be taken after I moved from Chicago to Princeton, and was a bit slowed up by the move. Fortunately several of my old Chicago friends moved to the Scripps Institution of Oceanography (SIO), previously in the case of Ed Goldberg and about the same time in the cases of Hans Suess and Harmon Craig. Gustaf Arrhenius of SIO had also already become a friend. So I had a good source for both samples and advice about where the

^{10}Be which had fallen in rain on the ocean would be found. My friends introduced me to deep-sea cores, red clay, globigerina ooze, and other oceanographic lore. I also made a useful visit to the Lamont Geophysical Observatory.

Since red-clay sediments show low sedimentation rates, and hence in principle should have high specific activity (decays per unit time per gram), I decided to try these first. Ed Goldberg kindly got me samples from two suitable cores, ranging from the sediment surface to depths of about a meter, and extending back in time on the order of a million years. The chemical separation was a bigger challenge, because of the large mass of sediment present, and the need in a first measurement to insure that all Be and all solids were dissolved. However, some improved separations had appeared in the literature, which made the processing of seven samples in a reasonable time (on the order of one per week) practical, though with chemical yields of only 30–40%. With the samples in hand, I needed a suitable low level beta-counting system.

My own shield and beta-counter system at Princeton were still under construction, so I was fortunate in having my friend Tom Sugihara fairly nearby at Clark University in Massachusetts. His department of defence grant had allowed him to receive the breech of a surplus cannon and reconfigure it as the shield for a counter system of just the right type. He willingly allowed me to use the apparatus over a long weekend. I took the train to Boston, borrowed my uncle's car, and drove to his university with the precious samples.

The first results were most encouraging. All but one sample with low yield showed clear beta signals. By Sunday morning I had completed the measurements, except for the necessity to measure an absorption curve for the 0.56 MeV beta. Many impurities, especially in the U and Th decay series, might have been responsible for the beta counts, but fortunately none of the likely contaminants had a similar energy. My problem was that I did not have with me a suitable set of foil absorbers.

I remembered Bill Libby's proverb that materials from the grocery store were cleaner than those from the scientific supply house, and also cheaper. The trouble was that in those days few

stores in the United States were open on Sunday. I drove around the small city of Worcester, and finally found a little shop open. Browsing the counters I attracted the owner's attention, even more when I could not tell him clearly what I wanted. Finally I found a polyethylene tablecloth which I guessed (correctly) had about the right thickness. He took my money, and warmly wished me success in my inexplicable use of it. His wishes certainly did no harm. A clean pair of lab scissors did the rest.

Attentive readers of my paper¹⁰ will note on examining the graph of the absorption curve that I plotted the comparison between the natural sample and a synthetic standard in an unusual way. The reason was my desire to conceal from critical (but not too critical) readers that the agreement between the natural sample and the standard was a good deal better than statistical errors would on average permit. Such extra-close agreement may of course (and did) happen by chance, but I have never been so tempted to 'fudge' data (by increasing the scatter) as here, in order to avoid suspicion. Interestingly, no one ever recognized my less improper device.

It was soon after this paper was submitted that Masatake Honda came from Bern to join, and transform, my research group. With him as a collaborator the difficult became easy, and the

impossible merely difficult. As one example among many, he developed the method, using the 'Honda column' by which my student John Merrill was able to isolate stable Be from sea water and measure its concentration of less than a part per trillion. This was a record low concentration for a stable species at the time. Early in this post-discovery period we acquired our own shield and counting system.

The correspondence between Bombay and Princeton had continued steadily, and we were able to help each other in a number of ways. I had suggested to Peters that if he chose we might publish back-to-back discovery papers, but he waved me on ahead, following not long after¹¹. (For an account of the independent discovery of ¹⁰Be by Peter's group, see the article by Lal, this issue.) Soon, when I moved to join my friends in La Jolla in 1958 at the start of UCSD's evolution into a science university, a member of the Peters group named Devendra Lal appeared almost at the same time. The collaboration among Honda, Lal, and myself has been one of the happiest and most fruitful of my life. While we in La Jolla turned our main efforts toward cosmic-ray produced activities in meteorites and later lunar materials, the group led by Lal, in Bombay and later in Ahmedabad, carried on the study of ¹⁰Be in deep-sea sediments and manganese nodules, al-

most alone, until the advent of accelerator mass spectrometry in the late 1970's made ¹⁰Be studies accessible to a much wider group^{12,13}.

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Recollections of an experiment on the charge composition of cosmic rays

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A 1968 project to look for relativistic nuclei in cosmic rays combined science and adventure.

In the spring of 1967 Professor Peters invited me to come to Denmark and spend a year working at the Danish Space Research Institute of which he was the director. We were old friends from Berkeley and it had been five years since we had worked together at the Niels Bohr Institute, so I was eager to work again with him and to take part in what sounded like a very interesting project: to build a balloon-borne instrument capable of measuring electronically the intensity of relativistic nuclei

in the primary cosmic radiation, identifying individual elements from lithium to iron, and of sufficient size to provide statistically significant data. As he had played such a pivotal role in the discovery of heavy nuclei in the cosmic radiation two decades earlier it seemed most appropriate that his institute would be undertaking this project.

By the end of September, with my wife and younger daughter, I arrived in Lyngby for what turned out to be a two-year stay. It was, indeed, an interes-

ting and successful project and I would like to recall, for the non-specialist reader, something about the experiment and how we carried it out. It is a personal account and subject to the tricks that memory plays after twenty-two years.

By the time I arrived at the institute the basic design for the cosmic-ray telescope was already well advanced and some prototype components were made. While this was not their only project, it was the first major cosmic-ray