

THE INTERACTIONS OF ATMOSPHERIC COSMOGENIC  
RADIONUCLIDES WITH SPACECRAFT SURFACES

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## SUMMARY

The discovery of the cosmogenic radionuclide  $^7\text{Be}$  on the front surface (and the front surface only) of the LDEF spacecraft (ref. 1) has opened opportunities to investigate new phenomena in several disciplines of space science. Our experiments have shown that the  $^7\text{Be}$  found was concentrated in a thin surface layer of spacecraft material. We are able to explain our results only if the source of the isotope is the atmosphere through which the spacecraft passed. We should expect that the uptake of beryllium in such circumstances will depend on the chemical form of the Be and the chemical nature of the substrate. We have found that the observed concentration of  $^7\text{Be}$  does, in fact, differ between metal surfaces and organic surfaces such as PTFE (Teflon). We note however that (a) organic surfaces, even PTFE, are etched by the atomic oxygen found under these orbital conditions, and (b) the relative velocity of the species is  $8 \text{ km}^{-1}\text{s}$  relative to the surface and the interaction chemistry and physics may differ from the norm.

$^7\text{Be}$  is formed by spallation of O and N nuclei under cosmic ray proton bombardment. The principal source region is at altitudes of 12-15 km. While very small quantities are produced above 300km, the amount measured on LDEF was 3 to 4 orders of magnitude higher than expected from production at orbital altitude. The most reasonable explanation is that  $^7\text{Be}$  is rapidly transported from low altitudes by some unknown mechanism. The process must take place on a time scale similar to the half-life of the isotope (53 days).

Many other isotopes are produced by cosmic ray reactions, and some of these are suited to measurement by the extremely sensitive methods of accelerator mass spectrometry. We have begun a program to search for these and hope that such studies will provide new methods for studying vertical mixing in the upper atmosphere.

## INTRODUCTION

The LDEF spacecraft was launched by the space shuttle Challenger on 7 April 1984 into a nearly circular orbit with an inclination of  $28.5^\circ$  and an altitude of 480 km. It was retrieved by the space shuttle Columbia on 12 January 1990 at an altitude of 310 km. Because of its large mass, long space exposure and the wide variety of materials onboard, the LDEF provided a unique opportunity for induced radioactivity studies. These measurements are still in progress and will be reported elsewhere.

The LDEF spacecraft has a twelve-sided cylindrical aluminium structure, 9.1 m long by 4.3 m in diameter (see Fig. 1). Its structure consisted of an open grid to which were attached various experiment trays designed to measure the effects of long space exposure on spacecraft materials and components. Throughout its orbital lifetime, the spacecraft was passively stabilized about all three axes of rotation, allowing one end of the spacecraft to point always toward the Earth, and fixed leading and trailing with respect to the orbital motion.

After its return to the Kennedy Space Center, gamma ray spectra were obtained along each of the 12 sides of the spacecraft using a germanium detector array provided by the Naval Research Laboratory. The gamma-ray line at 478 keV from the radioactive decay of  $^7\text{Be}$  was observed to emanate strongly from the leading side of the spacecraft. (ref. 2) The weaker signal observed from the trailing side of the spacecraft was later traced to the gamma-ray flux from the leading surfaces after attenuation from passing through the body of the LDEF.

## EXPERIMENTAL MEASUREMENTS OF RADIOACTIVITY

Individual components were brought to the Marshall Space Flight Center to quantify the residual radioactivity on the LDEF. Much of the counting work was performed at other radiation laboratories around the country. The authors are particularly indebted to Dr. Charles Frederick of the TVA Western Area Radiation Laboratory, Muscle Shoals, Alabama for many of the Al clamp plate assays. A high-purity germanium detector inside a low-level background facility was used to obtain spectra of small aluminium and steel samples taken from the leading and trailing sides. In Figs 2 and 3, gamma-ray spectra of two identical aluminium plates and two steel trunnion end pieces taken from the leading and trailing sides of the spacecraft are shown. A clear  $^7\text{Be}$  gamma ray signal was seen on materials from the leading side, with little or no signal above background on the trailing side.

In Figure 4 the  $^7\text{Be}$  activities for aluminum tray-clamps taken from trays all round the LDEF are shown, clearly demonstrating the leading edge effect. While  $^7\text{Be}$  is also produced by spallation of Al nuclei in the spacecraft by cosmic rays, first order calculations have shown it to be barely measurable. Also the known anisotropy of the cosmic ray flux (the east-west effect) should have resulted in higher production on the rear (west-facing side) of the LDEF. Another isotope  $^{22}\text{Na}$ , produced by spallation of spacecraft Al, clearly shows higher activity on the trailing edge of the satellite. Figure 5 shows tray clamp activities of  $^{22}\text{Na}$  about twice as high on the trailing as on the leading edge, in agreement with the east-west anisotropy of the cosmic rays and trapped protons. This evidence clearly pointed to a source of  $^7\text{Be}$  in the atmosphere being swept up by the front surface of the spacecraft.

In Table 1, the measured number of  $^7\text{Be}$  atoms per unit area on various spacecraft surfaces is shown. The results are corrected to the retrieval date of 12 January 1990 and for the offset angle from the leading direction. The areal density for  $^7\text{Be}$  on the aluminium and steel is the same within the experimental uncertainty, and is apparently not a strong function of the type or surface condition of the metal. However, the Teflon thermal coating which was used on many LDEF experiment trays, has a density of  $^7\text{Be}$  an order of magnitude lower than that found on the aluminum surface. The reason for this apparent difference in uptake efficiency is unknown, but could be related to the covalent-bond structure of the material. The explanation may be complicated, also, by the observed erosion of the Teflon surface by atomic oxygen.

TABLE 1  
LDEF Be-7 Surface Concentrations\*

Material	Be-7 Areal Density (x 10e5 atoms/cm <sup>2</sup> )
Stainless steel trunnion face	5.3 +- 0.7
Polished aluminum plate- Exp. A0114	6.7 +- 1.0
Anodized aluminum experiment tray clamp	4.6 +- 0.5
Teflon thermal cover	0.9 +- 0.2

\* Corrected for decay since recovery and for surface orientation relative to spacecraft ram direction.

### $^7\text{Be}$ PRODUCTION, DECAY AND DYNAMICS IN THE ATMOSPHERE

The short-lived isotope  $^7\text{Be}$  was first detected in the atmosphere by Arnold and Al-Salih in 1955, (ref. 3) and later mapped by others as a function of altitude and latitude (ref. 4-8). It is produced in the atmosphere by high-energy cosmic-ray interactions with air as are other radioisotopes such as  $^{14}\text{C}$  and  $^3\text{H}$ . Once formed,  $^7\text{Be}$  ions are presumed to oxidize rapidly and attach to small aerosol particles, which provide a downward transport mechanism from peak production regions of the atmosphere (ref. 9-16). The primary removal process for  $^7\text{Be}$ , which occurs on a timescale comparable to its half-life, 53.2 days, is the washout of the aerosol-attached  $^7\text{Be}$  in rain water (ref. 3-6).

At a given latitude above ~20 km, the production rate of  $^7\text{Be}$  varies vertically and directly in proportion to the oxygen-nitrogen gas density. Peak production per unit volume occurs in the lower stratosphere, at 12-15 km, below which the cosmic-ray flux is substantially attenuated. At higher altitudes, the number of  $^7\text{Be}$  atoms produced per unit volume decreases rapidly, but the number of  $^7\text{Be}$  atoms produced per unit mass of air is essentially constant. Balloon and aircraft measurements (ref. 6, 15) are in approximate agreement with this, although few measurements extend much above the peak production altitudes.

From the measured densities of  $^7\text{Be}$  on LDEF surfaces and in making some simplifying assumptions, we can estimate the concentration of  $^7\text{Be}$  atoms per cm<sup>3</sup> of air at the LDEF orbital

altitude. Since the lifetime of LDEF is much greater than the mean lifetime of a  $^7\text{Be}$  atom, and ignoring changes in altitude over the last 6 months in orbit, we assume a steady state relationship between pick-up of  $^7\text{Be}$  and loss by decay:

$$\frac{dn}{dt} = 0 = -k n_{\text{eq}} + n^* v p_s$$

where:  $n$  is the density of  $^7\text{Be}$  atoms on the surface at time  $t$   
 $k$  is the first-order decay constant for  $^7\text{Be}$   
 $n_{\text{eq}}$  is the steady-state surface density of  $^7\text{Be}$  in atoms  $\text{cm}^{-2}$   
 $n^*$  is the concentration of  $^7\text{Be}$  atoms in orbital space (atoms  $\text{cm}^{-3}$ )  
 $v$  is the spacecraft velocity ( $\text{cm})(\text{s}^{-1})$   
 $p_s$  is the sticking probability of Be on a metal surface

for first order kinetics of radioactive decay:

$$k = \frac{\ln 2}{t_{1/2}}$$

where:  $t_{1/2}$  is the half life

Thus we have:

$$n_{\text{eq}} = n^* v t_{\text{mean}} p_s$$

where  $t_{\text{mean}} = \frac{t_{1/2}}{\ln 2} = 76.8$  days for  $^7\text{Be}$

From the measured value of  $n_{\text{eq}}$ , assuming  $p_s = 1$ ,

we have  $n^* = 1.2 \times 10^{-7} \text{ cm}^{-3}$  at 320km

or a relative concentration of  $3.8 \times 10^6$  atoms per gram of air. In the peak production region, below 20km, previous measurements (ref. 4-8) yield a concentration of 1000  $^7\text{Be}$  atoms per gram of air, or  $\sim 0.1$  atoms  $\text{cm}^{-3}$ , in agreement with a simple calculation using known values of the cosmic-ray flux and the production cross-section for the isotope. Thus, the measured concentration of  $^7\text{Be}$  per unit mass of air at 320km is three to four orders of magnitude greater than it would be if it had been produced at that altitude.

The simplest explanation is that Be is quickly transported upwards from regions of the atmosphere where its numerical concentration is much higher (but not its relative concentration with respect to oxygen and nitrogen). This transport must take place on time scales similar to or shorter than the radioactive half-life (53.2 days).

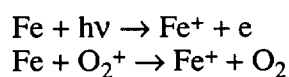
Vertical transport timescales at altitudes of several tens of km to 100km are considered to be too long to provide an efficient source, but Petty (ref. 17) has shown that above a certain altitude (not well defined, but about 100km) simple diffusion of the light nucleus in the Earth's gravitational field would provide an enrichment of a factor of 500 or more at 300km. Turbulent mixing below 100km cannot be easily invoked as it proceeds at times scales longer than the isotope half-life. More detailed calculations are needed to see if closer agreement can be reached.

## ATMOSPHERIC CHEMISTRY AND SURFACE CHEMISTRY OF Be

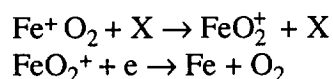
Thus far we have considered the chemical form of Be to be single atoms of mass 7. At low altitudes, rapid oxidation would be expected and in regions close to the tropopause, this would be followed by rapid absorption onto aerosol particles. The raining-out of these Be-bearing aerosols has proved a useful tool for measuring the efficiency of tropospheric mixing by thunderstorms.

If the Be were in the form of its normal oxide BeO (mass 23) at altitudes above 100 km, we can no longer rely on rapid diffusion to higher altitudes. While not much appears to be known of Be chemistry in the upper atmosphere, a great deal of work has been done on the chemistry of metals ablated into the upper atmosphere from meteorites. These metals include Mg, Ca, Al, Si and Fe.

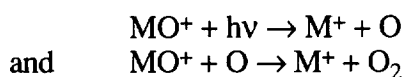
$^7\text{Be}$  is formed as a "hot" atom or ion, which must rapidly thermalize with the atmosphere. From studies of meteoritic ions in the atmosphere we may draw some general conclusion as to the chemical form in which the Be atom will finally take. The form of the meteoritic ions is highly variable with altitude and between day and night. Electropositive metals readily form positive ions:



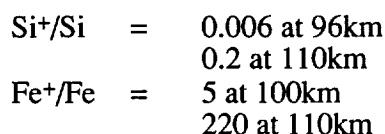
At low altitudes neutralization may occur (X is a third molecule):



In general at altitudes in excess of 100km the metal (M) oxides cannot survive in appreciable quantities due to reactions such as

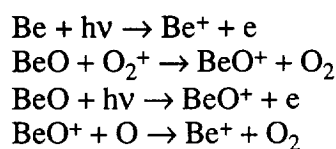


Thus at higher altitudes the singly-charged positive ion dominates for most metallic species studied. Of interest is the ratio  $\text{M}^+/\text{M}$  which varies with altitude and electropositive character of the metal. Examples of some measured ratios from the literature (ref. 18) for silicon and iron are:



Thus above 150km (and perhaps as low as 100km) most Be should exist as  $\text{Be}^+$ .

Important reactions might be:



It is reasonable that a positive metal ion striking a clean metal oxide surface, especially with several eV kinetic energy, should easily enter the oxide lattice and remain trapped. While most metal (and metal oxide) surfaces in the ground-level atmosphere are usually found (by ESCA techniques) to be covered with a layer of hydrocarbon contamination, this is not the case with the leading surfaces of LDEF which are known to be continuously cleaned of combustible material by the action of atomic oxygen in the atmosphere. These atomic oxygen/satellite surface interactions have been intensively studied on the LDEF. The interaction of Be ions with metal oxides is an example of a new kind of chemical reaction between atmospheric species and satellite surfaces and has implications yet to be explored.

We investigated the form of binding of the Be to the aluminium surfaces on LDEF. Possibilities included (1) binding within an adsorbed contaminant layer, for example of hydrocarbon; (2) binding of Be-containing particulates, perhaps aerosols or meteoritic debris and (3) binding within the native oxide found on aluminum and other metals. Two kinds of Al plates from the LDEF were measured, some with several microns of oxide produced by anodization and the second type a polished Al plate from the UAH Atomic Oxygen Experiment A0114 (ref. 19). The oxide on this was only expected (ref.20) to be 50 - 100 Å thick.

The polished Al plate was coated with a solution of Collodion, which was then dried, stripped off and counted. No Be activity could be associated with the Collodion film. The method is used in industry to reliably and quantitatively remove particulates from sensitive surfaces. Next the plate was wiped first with alcohol, then with xylene. No activity was removed with the wipes. Finally an acid etch was used to remove the top 10 microns of the surface. The etch solution contained most of the Be activity formerly on the plate: that remaining being associated with either unetched surface or with re-adsorption of Be<sup>2+</sup> ions onto the Al. This might be expected since a stable Be carrier solution was not used. The experimental results are consistent with the hypothesis that the Be species were penetrating the aluminium oxide layer on the surface of the plates and becoming permanently fixed in the oxide lattice. We believe the penetration to be of the order of one nanometer, since the kinetic energy of the Be species relative to the spacecraft was only 2.5eV. We do not have the capability to remove such a thin layer from large areas of metal surfaces, and thus cannot measure a depth/composition profile for the species.

<sup>7</sup>Be is not the only nucleus produced by cosmic rays in the atmosphere. In fact all stable nuclei of lesser atomic weight than oxygen, nitrogen and argon must be formed. The means to detect the extremely small concentrations of most of these nuclides (in the presence of naturally occurring levels) do not exist. A few other unstable nuclides exist however with half-lives long enough to allow measurement, and short enough that there is no other natural background concentration. These are <sup>14</sup>C, <sup>10</sup>Be and possibly <sup>26</sup>Al (from argon).

The only method sensitive enough to measure these nuclides is accelerator mass spectrometry (AMS) (ref. 21). While the method has proved most useful for radioactive nuclei, emission of radiation by decaying nuclei is irrelevant to the AMS technique. Rather, all atoms of the nuclide are counted in the mass-spectrometer, giving some major advantages over radiation-counting methods.

<sup>10</sup>Be is produced in a similar manner to <sup>7</sup>Be, by spallation of N and O induced by secondary neutrons from cosmic ray interactions in the atmosphere. The production efficiency is about 0.5 that of <sup>7</sup>Be, however its half-life is 1.5 x 10<sup>6</sup> yrs (compared with 53.2d for <sup>7</sup>Be), resulting in measured ratios <sup>10</sup>Be/<sup>7</sup>Be of about 3 in the stratosphere (ref. 22). While the atmospheric chemistry of the two isotopes should not differ appreciably, the diffusion of neutral atoms to higher altitudes should show measurable differences because of atomic mass.

<sup>10</sup>Be decays to <sup>10</sup>B by internal conversion, emitting electrons over a wide energy range, while <sup>7</sup>Be decays to <sup>7</sup>Li by electron-capture, emitting gamma-rays of very narrow energy

distribution. The latter allows rates of a few decays per day to be measured in our low-level counting apparatus, while the former poses insurmountable counting problems. AMS however can detect  $^{10}\text{Be}$  with undiminished sensitivity. We are currently working on chemical separation techniques\* and plan a  $^{10}\text{Be}$  run at the University of Pennsylvania† in fall of 1991.

We also plan a search for another cosmogenic radioisotope,  $^{14}\text{C}$ , also using AMS. We plan to use the NSF-Arizona facility†† to investigate the take up of  $^{14}\text{C}$  species by blanket material from LDEF. Carbon chemistry is completely different from that of the metals. Cosmogenic carbon should form CO and  $\text{CO}_2$  rapidly in the lower atmosphere but its behavior at higher altitudes is unknown. Upwards diffusion of the oxide species would not be favored (their masses are 28 and 44) and the adsorption on spacecraft materials is unknown.

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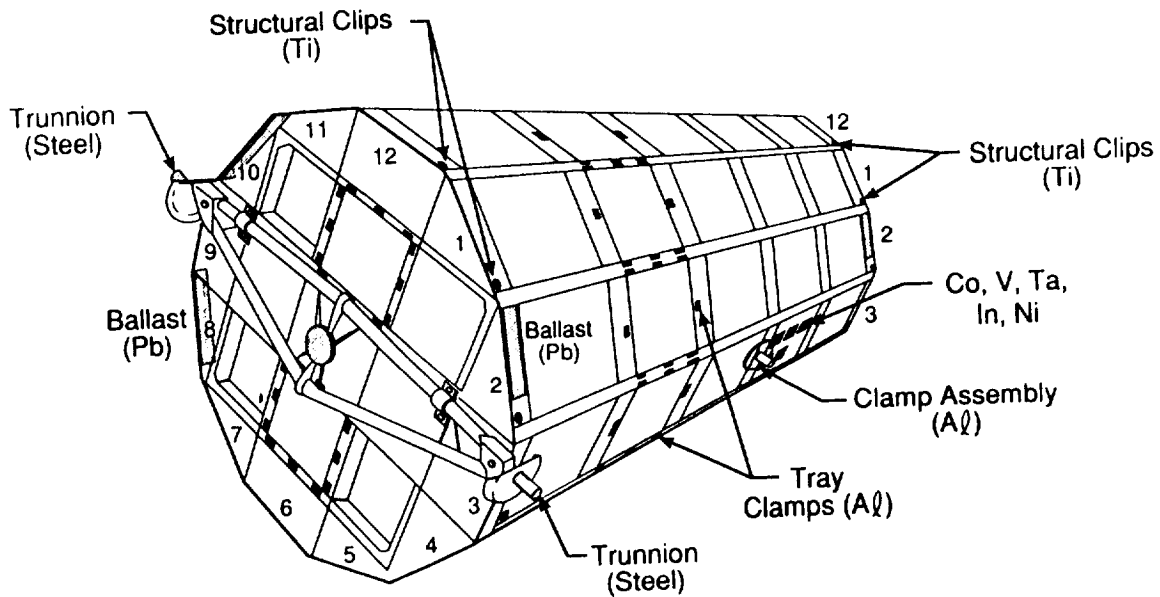


Figure 1. The LDEF spacecraft, showing the location of pieces of material studied for induced radio-activity

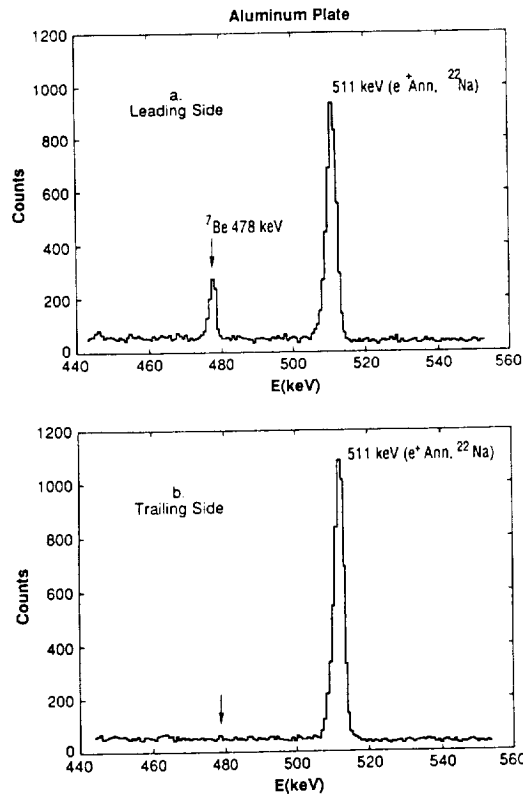


Figure 2. Portion of the gamma-ray spectrum obtained from an aluminum plate (a) on the leading side and (b) on the trailing side of the LDEF. The  ${}^7\text{Be}$  line at 478 keV is seen only on the leading side.

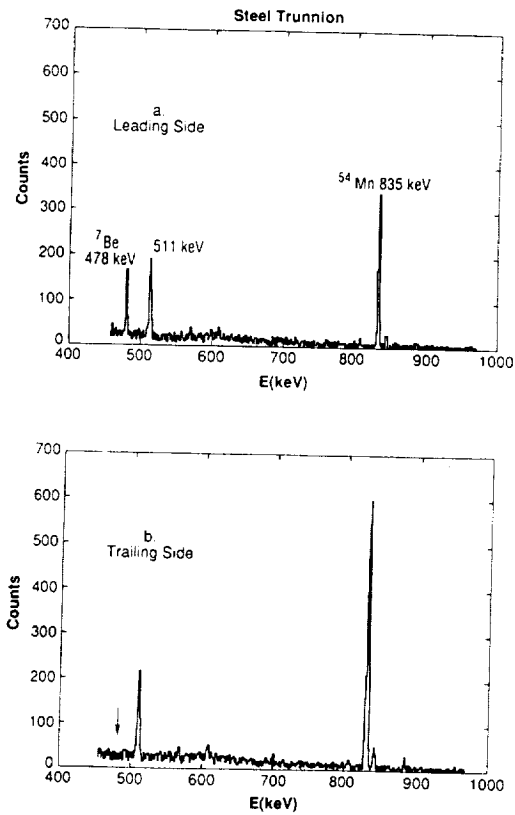


Figure 3. Portion of the gamma-ray spectrum taken from the stainless steel trunnion (a) on the leading side and (b) on the trailing side of the LDEF. The  ${}^7\text{Be}$  line is seen only on the leading side, whereas the spallation products produced within the steel itself,  ${}^{54}\text{Mn}$  and  ${}^{22}\text{Na}$  are seen on both trunnions.

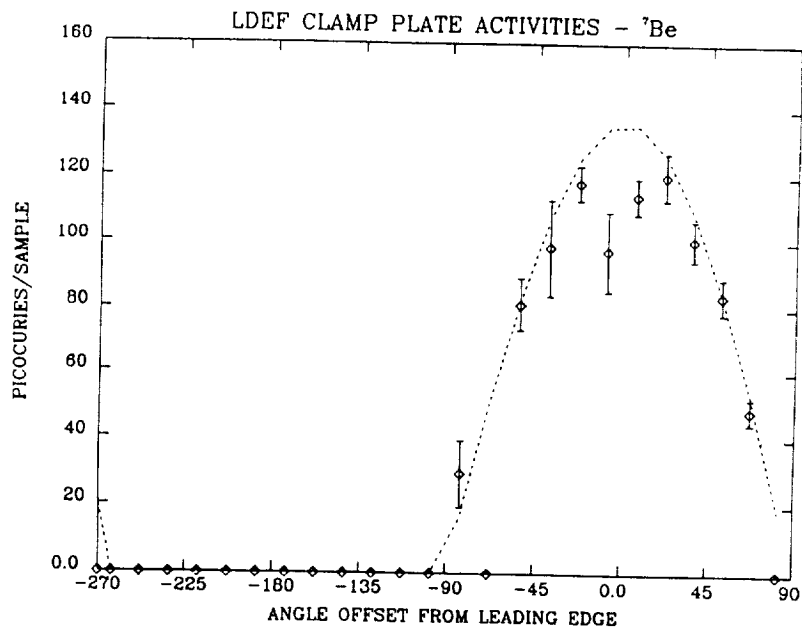
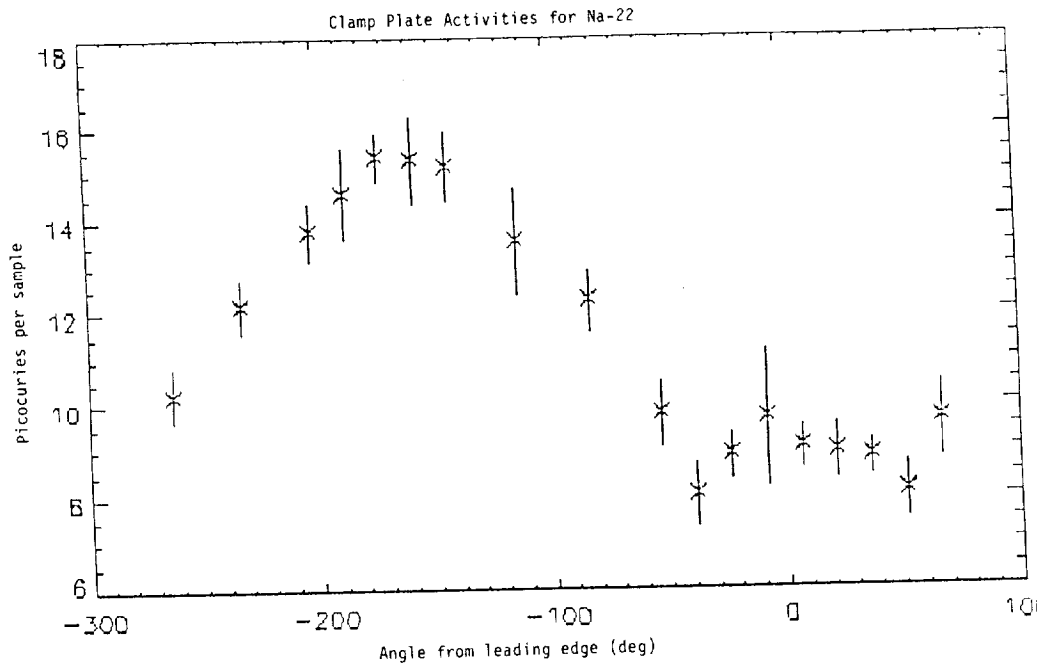


Figure 4.  ${}^7\text{Be}$  activities for aluminum tray-clamps taken from all round the LDEF. The leading edge is nominally 0 deg and the trailing edge 180 deg.  ${}^7\text{Be}$  activity is clearly a function of surface area projected in the forward direction of the spacecraft.



**Figure 5.**  $^{22}\text{Na}$  activities for aluminum tray clamps taken from around the LDEF. The leading edge is 0 deg. and the trailing edge 180 deg. Activity is peaked at the trailing edge but found all round the spacecraft. As expected from the anisotropic cosmic ray and trapped proton fluxes, more activity is induced in materials on the westerly (trailing) side of the spacecraft.

