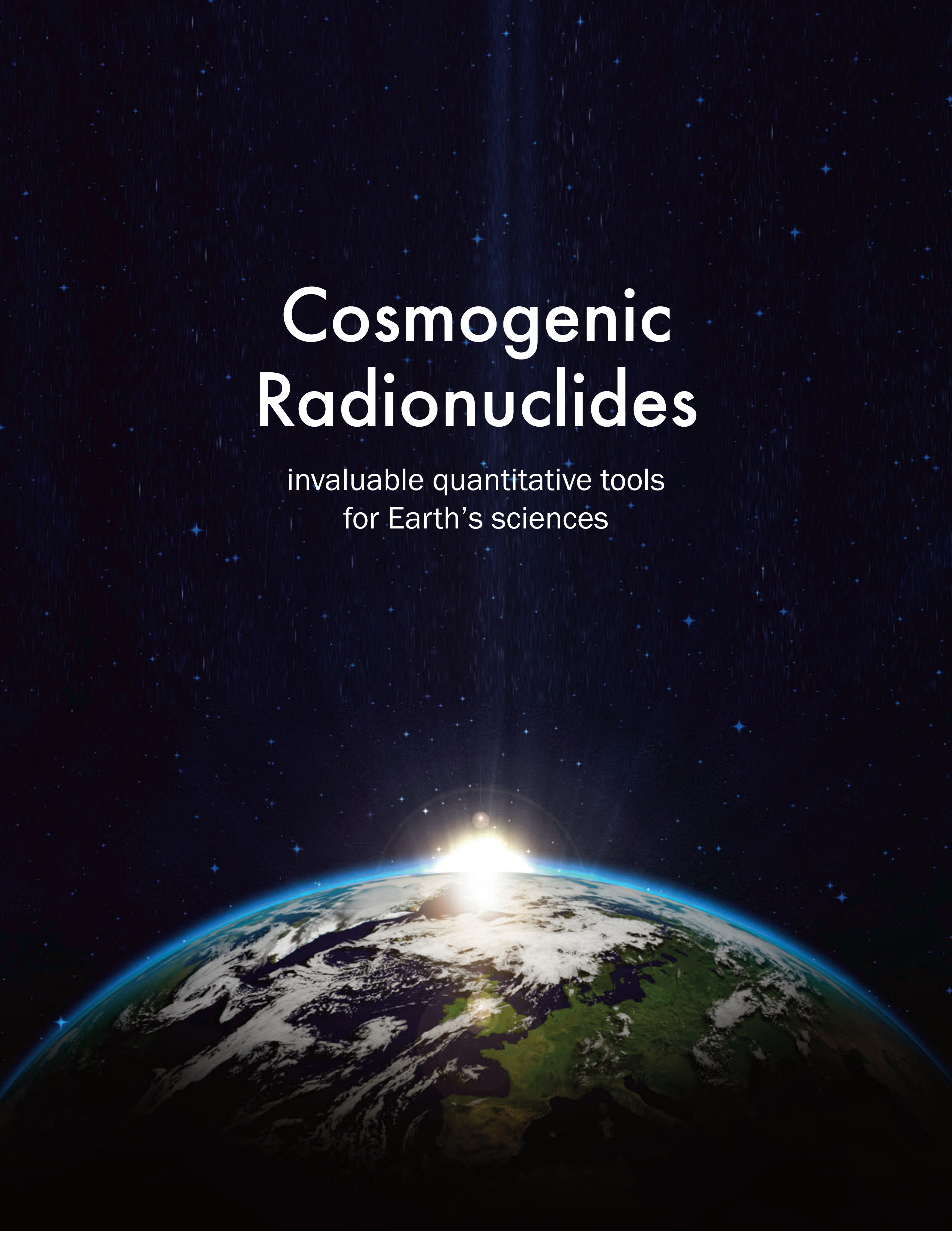

Cosmogenic Radionuclides

invaluable quantitative tools
for Earth's sciences

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Accélérateur pour les Sciences de la Terre, Environnement, Risques
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Cosmogenic radionuclides: invaluable quantitative tools for Earth's sciences.

The production of cosmogenic radionuclides on Earth result from nuclear reactions initiated by primary and secondary energetic cosmic ray particles which, during their passage through the atmosphere and into the first few meters of the Earth's crust, interact with various atoms of atmospheric gases and surface rocks, respectively. In this way there are two types of cosmogenic produced radionuclides – the atmospherically produced cosmogenic nuclides which mainly are ^{10}Be and ^{14}C, and the *in situ*-produced cosmogenic nuclides (produced within minerals of the Earth's crust) with ^{10}Be, ^{26}Al and ^{36}Cl being the more commonly used isotopes. The former production is far more intense because cosmic rays dissipate most of their energy in the nuclear reactions occurring in the Earth's atmosphere, and only a small (~0.1%) quantity of cosmic ray derived particles reach the Earth's surface with sufficient energy to induce an effective nuclear reaction. For atmospheric production, the target atoms are N and O molecules, whilst for terrestrial in-situ production it is O and Si in quartz and Ca and K in other mineral rocks. The half-lives for the above mentioned radionuclides range from 5,730 years to 1.39 million years.

1: Atmospherically produced cosmogenic radionuclides

Mainly produced in the atmosphere through nuclear reactions (spallation reactions) on oxygen (O) and nitrogen (N), the particle-reactive ^{10}Be is rapidly transferred to the Earth's surface in soluble form by precipitation where it is ultimately removed from water on settling particles and either deposited in marine and lacustrine sediments, or efficiently retained onto continental sediment components where it decays with a half-life of 1.39 ± 0.01 Ma.

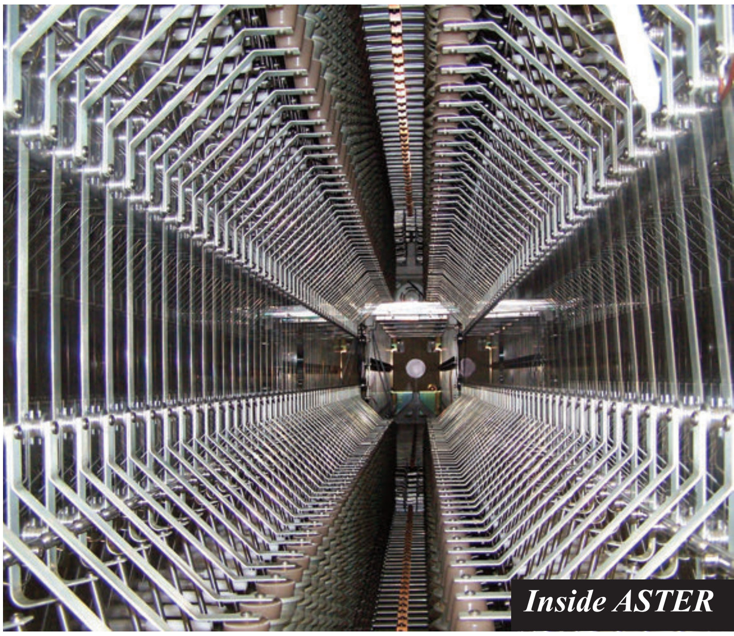
Considering the precision and the detection limits of the analytical methods which have been developed to count the small concentrations of these long lived radionuclides (see below), this opens up the possibility to date various land and marine based sedimentary deposits in the range of 0.2 to 14 Ma.

However, because the incoming primary cosmic ray particles are electrically charged particles (they are 96% protons and only 4% alpha particles), their intensity is modulated by both variations in the strength of Earth's geomagnetic dipole moment and by the solar wind which modulates the Earth's magnetosphere. As the production rate of cosmogenic ^{10}Be is directly proportional to the cosmic ray flux (which has changed over time), it is also inversely proportional to the geomagnetic field dipole moment. Therefore concentration enhancements (decreases) in ^{10}Be per gram sediment are theoretically recorded in marine cores at the same stratigraphic level as the RPI (Relative PaleoIntensity) lows (highs), taking in account the time delay due to the lock-in of the post-depositional remanent magnetisation.

In order to properly interpret an atmospherically-produced ^{10}Be concentration, it is critical to understand that

The french AMS national facility ASTER

the absolute ^{10}Be concentration is also controlled by various environmental conditions and factors, such as fallout rate, sediment scavenging efficiency, the specific surface of the settling sedimentary particles, and the chemical and grain size composition of the sampled sediment. To account for this dependency, the stable isotope ^{9}Be is used as a normalising parameter for ^{10}Be as long as the two isotopes are extracted from the same geochemical phase. However there are fundamental differences in the input functions and sources of both beryllium isotopes, i.e. atmospheric ^{10}Be is predominately in a dissolved phase whilst the bulk of stable ^{9}Be is introduced by detrital and particulate inputs. Thus the parameter of significance is ultimately associated with the authigenic phases of the sediments – i.e. the fraction due to the adsorption onto particles of both the dissolved ^{9}Be and ^{10}Be. Sequential leaching experiments have thus been developed to selectively extract this authigenic phase from the pertinent sedimentary deposits and measure their ^{10}Be/^{9}Be ratio. Studies up to now exclusively performed on marine sediments have effectively demonstrated that the ^{10}Be/^{9}Be ratio measured in this authigenic phase represents the ^{10}Be/^{9}Be ratio of soluble Be at the time of deposition. Providing that (a) the initial concentration relative to the normalising element (the initial ratio) can be accurately estimated, (b) that the selected samples have remained “a closed system ” (i.e. no exchange of the cosmogenic isotope and its normalising element), then the classical ‘dating’ equation $N_{(t)} = N_0 e^{-\lambda t}$ (where $N_{(t)}$ is the measured ratio; N_0 the initial ratio; λ the radioactive decay constant and t the age of the sediments) can be applied to the measured authigenic ^{10}Be/^{9}Be ratios to calculate the ages of the studied sediments.



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The question now remains how to determine the initial ^{10}Be/^{9}Be authigenic ratio? This is readily available by measuring authigenic ^{10}Be/^{9}Be ratios in recent samples deposited in similar environmental and geochemical settings to establish the regional initial authigenic ^{10}Be/^{9}Be ratio. This value serves to calculate the absolute age of the samples following the radioactive decay equation described above. It nevertheless varies through time due to fluctuations in ^{10}Be production rates associated with shifts in the strength of the geomagnetic field which affects the accuracy of the calculated ages.

Tectonic fault traces in Mongolia

1.1: Production rate variabilities and geochronological calibration

The consequences of these geomagnetic intensity variations on the ^{10}Be production rate may be quantified from rapidly accumulating marine sediment cores. Over the past twenty years, several studies have indeed shown that the ^{10}Be production rate is, as theoretically expected, anti-correlated with changes in paleointensity. These studies have mainly focussed on cores which span the last ~50 ka that includes the well-known peak in geomagnetic field called the Laschamp excursion, which occurs together with a large drop in ^{10}Be concentration. Their internal consistency, in contrast to the records of relative paleointensity obtained for the same period, clearly indicates the strong potential of combined paleomagnetic and ^{10}Be concentration studies to accurately document the geomagnetic field and ^{10}Be production rate changes in the past.

Systematic study of the polarity and the geomagnetic field relative paleointensity coupled to that of the atmospheric ^{10}Be concentration in marine sediments with high sedimentation rates allows: 1/to better constrain the impact of variations in the intensity of the geomagnetic field on the production rates of other cosmogenic nuclides, especially on the production rate of ^{14}C; 2 / to better understand the internal mechanisms and constrain the origin of the geomagnetic field and especially of it numerous disturbances (excursions, reversals).

1.2: Geochronology and tracing

Atmospherically-produced cosmogenic nuclides are used to date marine and continental sedimentary records through the highlighting and characterisation of remarkable events generating global simultaneous significant atmospheric cosmogenic nuclide concentration variations in these reservoirs, including polar ice cores, which also allow their chronostratigraphic correlation.

In addition, atmospherically-produced cosmogenic nuclides are univocal tracers of the participation of sedimentary deposits to different geodynamic processes such as, for example, recycling of crustal material in subduction zones, the leaching of sedimentary lenses by hydrothermal fluids, the incorporation of sedimentary material in basalts.

Inventory of these cosmogenic nuclides along sedimentary deposit profiles compared with theoretical results calculated from annual flows at the sampling sites allows precising the modes of implementation, development and evolution of soils.

1.3: Archaeometry

Finally, atmospherically-produced ^{10}Be has been used to absolutely date continental fossiliferous deposits over the last 10 Ma.

2: In situ-produced cosmogenic radionuclides

In situ-produced cosmogenic nuclides result from the penetrating secondary cosmic ray flux bombarding surface rocks and the shallow few meters of the lithosphere. Similarly as for atmospheric production, in-situ production rates also depend on energy-dependent cross sections and on the cosmic ray flux reaching the Earth's surface. *In situ*-production thus also depends on the strength of the Earth's magnetic field, which results in a latitudinal variation, whilst an altitudinal variation in production rate is a result of dissipation of the cosmic radiation within the Earth's atmosphere.

Due to the efficient dissipation of their energy through nuclear reactions, the flux of the nuclear active particles, and therefore the cosmogenic nuclide production rates, decrease exponentially with the mass of overlying material with a characteristic attenuation length Λ (g.cm^{-2}). Two main types of secondary particles (neutrons and muons) induce *in situ*-production in the lithosphere. The effective production attenuation length of neutrons is short (~130-180 g.cm^{-2}) relative to that of muons (>3500 g.cm^{-2}). This means that whereas neutron-induced production is dominant near the surface, below a few meters reactions with muons become dominant. To be applicable to a broad range of sampling sites, a procedure for determining production rates for more complex geometries of irradiation is required. Finally, since the half-lives of ^{10}Be, ^{26}Al and ^{36}Cl are very short compared to the Earth's age, their primordial component has vanished.

For surface undergoing denudation, if we assume that the studied rock has undergone a single cosmic ray exposure episode, the cosmogenic nuclide contents increase with time until they reach a steady-state bal-

ance between production and losses due to denudation and radioactive decay. The measured concentrations may then allow estimating either minimum exposure ages neglecting denudation or maximum denudation rates assuming exposure times long enough to reach the steady-state balance concentrations.

Along a depth profile, because of the significantly different attenuation length associated with muons and neutrons, separate determination of ^{10}Be produced by each of the two mechanisms theoretically offers the opportunity to estimate both the exposure age and the denudation rate of the surfaces affected by relatively constant denudation rates. Because cosmogenic nuclides produced by neutron-induced spallation reach steady-state with respect to denudation loss more rapidly than those resulting from muon-induced reactions, ^{10}Be produced at the surface may be used to estimate the denudation rate and that produced by muons at several meters depth to estimate the exposure time.

2.1: Tectonics and paleoclimate

The use of *in situ*-produced cosmogenic nuclides for dating geomorphic markers such as alluvial fan surfaces, channels, glacial moraines, fault scarps has been developed. These dated markers then allow establishing the chronology of continental climatic events and, when offset by tectonic activity, constraining slip rates and thus to better constrain the seismic hazard.

2.2: Gravitational movements

In situ-produced cosmogenic nuclides allow studying the initiation and evolution of different types of gravitational movements in alpine regions. The provided chronological constraints allow determining the mechanisms involved in their initiation, their past evolution and thus to predict their future evolution.

2.3: Weathering, denudation

They also have been developed for a detailed and transcontinental understanding of the processes involved

A tectonic fault scarp in Italy (Maratea)

in the emplacement and the development of various types of weathering profiles. In particular, they are now widely used to determine pre-anthropogenic denudation rates at the scale of watersheds.

Environmental changes may initiate a process of burial on a surface which is undergoing steady state denudation (i.e. sediment transfer to and deposition on an accumulating surface). Under these conditions, loss of cosmogenic nuclides due to denudation ceases and the production rate of buried sediment decreases with depth compared to that of the original surface. Depending on the sediment accumulation rate, subsurface concentrations of cosmogenic nuclides may increase in a sediment profile undergoing burial as long as it remains close enough to the surface so production outweighs radioactive decay. This implies that the evolution of the ^{10}Be as a function of depth for an accumulating sediment profile (allochtonous processes) is fundamentally different from that for a profile resulting from *in situ* weathering mechanisms (autochthonous processes). On the other hand, the burial of a previously emplaced profile may result in exponentially decreasing ^{10}Be concentrations that are significantly higher for their present depth than those allowed in the zero denudation rate autochthonous scenario. The study of depth profiles of ^{10}Be thus allows to distinguish between different surface emplacement scenarios.

2.4: Archaeometry

When field observations or previous studies suggest a complex exposure history for the sampled bedrock or boulder, the concentration ratio of ^{26}Al/^{10}Be in the same rock may be used to determine if that sample has undergone a prolonged period of deep burial. The significantly different radioactive decay constants for ^{10}Be and ^{26}Al implies that in the case of any denudating surface, the ^{26}Al/^{10}Be ratio can only evolve within a closed region defined as the « steady-state denudation island ». A ^{26}Al/^{10}Be ratio that measures below this

An alpine landslide in France (Lauvitel)

“island’ will necessarily be the result of a partial or total burial time, indicating a more complex exposure history for that sample, which would otherwise go undetected had only one radionuclide been measured. Although mathematically there are an infinite number of pathways of cyclic burial and exposures periods that can produce a depressed ^{26}Al/^{10}Be ratio, one can estimate a minimum burial time and an initial ^{10}Be concentration (or steady state denudation rate). This method had been used worldwide to absolutely date cave sediments as well as their containing hominin artefacts.

3: Accelerator Mass Spectrometry

All the applications listed above would have not been possible without the development since the 1980s of the Accelerator Mass Spectrometry (AMS) technique which adds to “classic” electrical and magnetic selections, which allow for the selection of ions with given charges and mass (Mass Spectrometry), an acceleration stage (by Accelerator) which makes possible the elimination of molecular interference during high energy dissociations, thanks to the energy provided, and subsequently allows access to the nuclear structure of the selected ions and their analysis according to atomic number. They can be differentiated in this way from their natural isobars which are at least 10^8 more abundant.

One of the most remarkable breakthroughs linked to the development of this method of analysis is the improvement by a factor of 10^6 to 10^9 of the sensitivity of detection relative to more classic techniques, in particular those based on radioactive disintegration counting. This technique is consequently particularly well-adapted to the study of cosmogenic nuclides, which are extremely rare in natural environments, and which are concealed by interference from much more abundant isobar atoms. Indeed, for the most commonly used ones (^{10}Be, ^{26}Al, ^{36}Cl), the threshold for detection by AMS is to the order of only a few tens of thousands of atoms. It thus becomes possible:

- to work with much smaller quantities of matter than are needed for other techniques,

- to date more different types of objects on a much broader temporal scale than was previously available,

A glacial erratic boulder

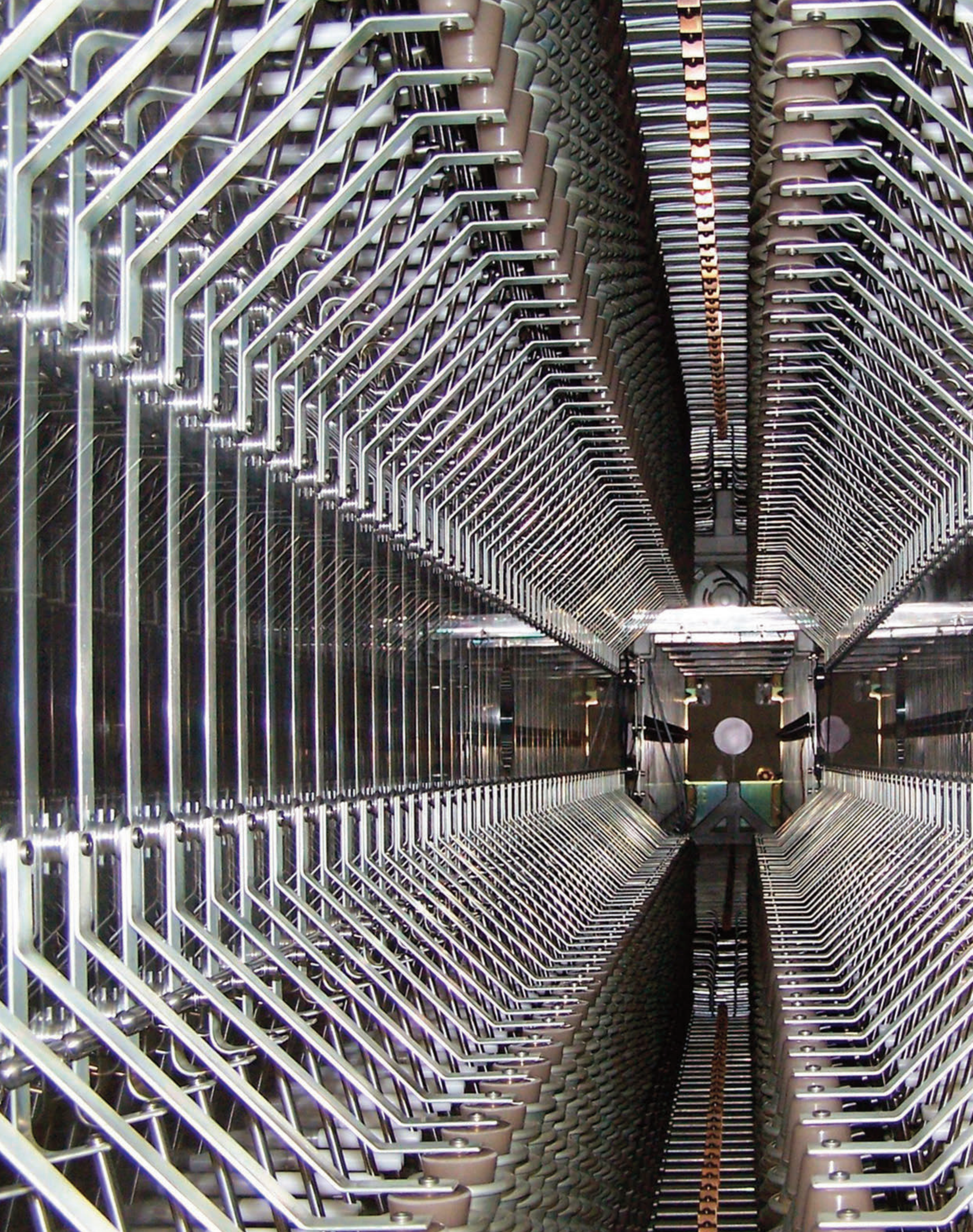
- to considerably reduce the time required to acquire the data,

- to very noticeably simplify the physical and chemical manipulations required when preparing samples for analysis.

4: Conclusions

Thus, since the advent of the AMS technique, quantitative information generated by cosmogenic nuclides produced in the atmosphere have become crucial to the comprehension of the interactions, both between the different compartments of the terrestrial environment and between these same compartments and the extraterrestrial environment. The atmospheric component of the radioactive cosmogenic nuclides is also a potential tool for absolute dating of the reservoirs in which they accumulate. ^{10}Be with its period of ~$1.4.10^{6}$ years appears, in particular, to be especially well-adapted to determine chronology of both polar and continental glacial accumulations as well as marine and continental sediment records over the last 14 million years. In this last case, the accessible radiochronological data may have fundamental implications in fields of research as significant as paleo-anthropology, paleo-climatology, and paleo-magnetism.

What is more, the continual lowering of detection limits over the past decade as a result of technological and methodological advances has made possible measurements of the cosmogenic nuclide (^{10}Be, ^{26}Al and ^{36}Cl, mainly) concentrations produced directly in the Earth's crust (in-situ production) and has thus allowed quantitative investigations previously impossible. These investigations concern, more specifically, 1) superficial deformations, thus permitting the analysis of certain natural hazards such as seismic and gravitational events, 2) continental paleo-climatology and in particular, the chronology of the most recent deglaciations, 3) interactions between erosion – tectonics – climate, 4) processes of development and evolution of continental surfaces, 5) developments of alteration profiles, and 6) estimations of denudation rates on the scale of the watershed.



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