Using of $^{53}$Mn and $^{10}$Be Cosmogenic Isotopes for Geochronology and Monitoring of Cosmic Rays in Terrestrial Rocks

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To cite this article:

Received: December 2, 2016; Accepted: December 26, 2016; Published: January 19, 2017

Abstract: The article discusses the methods of measuring the accumulation of radioactive isotopes $^{53}$Mn (with a half-life of $T = 3.7$ million years) and $^{10}$Be ($T = 1.6$ million years) in the iron-containing rocks. Knowledge of accumulation dynamics of these two isotopes would allow knowing both the time of shielding and flux of cosmic rays and also changes assessing the variations in the intensity of cosmic rays, the timing of glaciations, geological changes, and climatic processes in the world in retrospect of 0.1-10 million years. The main attendance paying to neutron - activation method of isotope analysis as low-cost both in money and time and more adapted for numerous sample processing. The advantages of isotope measurements in lunar craters are emphasized.

Keywords: Cosmic Rays, Cosmic Rays Variations, Radioactive Isotopes, Isotopes Accumulation, Geological Changes

1. Introduction

It is known that on the Earth there exist natural archives of cosmic particles and radiation, having a good memory about astronomical phenomena on a large scale in the past time. These are the tree rings, corals, polar ice, sediments of seas and oceans, stalactites, etc.

Cosmic rays (CR) generated by the natural particle accelerators in space, continuously bombard the Earth's surface and produces the radioactive elements, in particular, $^{53}$Mn (with a half-life $T = 3.7$ million years) and $^{10}$Be ($T = 1.6$ million years) in the iron rocks. These radioactive elements are very convenient for geochronology goals in the time interval of up to 10 million years.

In this paper we propose to measure the accumulation of these isotopes in the rocks inside the area of interest. The quantity of isotopes formed allow to estimate the time of their accumulation and periods of glaciation and geological bedding planes, which interrupted the process of isotopes production due to shielding by layers of ice, water and volcanic rocks. These data are extremely valuable for creating historical maps of glacial and geological changes in different parts of the Earth. Knowledge of the accumulation dynamics for these two isotopes apart from their accumulation time allows determining also the intensity of the cosmic rays and their possible variations in the past that it is very important for the understanding of climate change and periods of species mass extinctions and preventing the recurrence of similar processes in the future.

Impact of rocks erosion can be account for as follows. The nuclear cascades in the ground have a specific profile of $^{53}$Mn ($^{10}$Be) accumulation density in depth. Typical cascade length is about 100 g/cm$^2$, and depends on the rocks chemical composition.

It is supposed to take samples of very hard rocks such as granite or basalt to form cores of 1-1.5 meters long and measuring the density of cosmogenic products in the cross sections in steps of about 10 cm. In the presence of erosion the density profile along the length will be broken with respect to that in the absence of erosion. Measurement of profile parameters will allow an amendment to the erosion.

Similarly, the accumulation of radioisotopes occurred in meteorites that fell to Earth's surface. Estimation of radioisotopes accumulation in meteorites is an independent test of CR intensity and their variations over time. Earlier work [1] by means of the Monte – Carlo method the
production rate of cosmogenic isotopes $^{22}$Na, $^{36}$Cl, $^{40}$K, and $^{41}$V in iron meteorites was calculated. $^{40}$K isotope with a half-life of $1.3 \times 10^9$ years is an indicator of long-term CR variations associated with the evolution of the Galaxy and the problem of the origin of the CR itself. From a comparison of $^{40}$K,$^{36}$Cl activity ratio for meteorites with the ages of about 1 billion years with the corresponding calculated ratio for the current CR intensity it has been concluded [2-3], that the galactic cosmic ray intensity (GCR) in the past was 2.7 times lower than today. On the contrary, the data of [1] show the equality of the intensities of the GCR billion years ago and today.

Investigation of the possibility of cosmic rays generation by the explosion of supernovae associated with the investigation of GCR variations on a time interval of up to 100 thousand years. To change the average energy density of cosmic rays (1 eV/cm$^2$) so that this change can be measured requires a supernova explosion at the close of the solar system distance: less than 100 pc (distance of 1 pc light passes over 3 years). It is known that in the area with a radius of 100 pc average burst should occur once every 100 thousand years, that is a possibility of explosion in real time is very small. Moreover, even if such an explosion took place, then to establish the source nature (supernova or not) it is necessary to conduct continuous measurements required for tens or thousands of years.

Method of cosmogenic isotopes, mentioned above, allows reregistering the cosmic rays from a supernova explosion in the distant past.

The first experimental results on the content of $^{10}$Be in polar ice over the last 40 thousand years were published in the early 80s [4]. A significant increase in the intensity of the cosmic ray has been found in the time interval of about 10-40 thousand years ago. The result obtained revealed a quantitative and qualitative evidence of a supernova explosion near the solar system at a distance of no more than 50 pc of the Sun. Subsequently, additional experimental data on the $^{14}$C, $^{10}$Be and $^{36}$Cl fully confirmed the conclusion of the explosion of a supernova in the vicinity of the Sun about 35 thousand years ago; please see Fig. 1 taken from [5].

Thus, the above data justify the need for research in the time interval 0.1-10 million years ago to search for variations of GCR.

A study of the isotopic ratio of $^{10}$Be/$^{53}$Mn is of great interest to both the stony-iron meteorites, and for terrestrial rocks, which are not covered by sediments and not exposed to significant weathering in the past few million years. This ratio is sensitive to the significant variations in the flux of cosmic rays in the near-Sun and near-Earth space in the range of several million years.

2. Benefits of $^{53}$Mn in Geochronology

Currently the technique for measuring of $^{10}$Be, which is formed by cosmic rays in the spallation reactions, is well established. The main elements of the rock-forming crust such as O, Si, and Al are used as the target in this case.

Using another long-lived isotope such as $^{53}$Mn extends the timeline available for the measurement. An analysis of the accumulation of two long-lived isotopes $^{10}$Be and $^{53}$Mn with different in 2 times lifetime allows to evaluate the CR flux in past epochs ($10^4$-$10^7$) years.

Under irradiation of cosmic rays protons the main formation channel of $^{53}$Mn is represented by the reaction:

$$^{56}{\text{Fe}}(p, \alpha)^{53}{\text{Mn}}.$$  \hspace{1cm} (1)

Taking into account the multiplicity of 4-5 at the energy of cosmic rays $E_p = 10^8$ eV the cross section of this reaction according to ENDF is $\sigma_{\text{p,}\alpha} = 0.4 \times 10^{-27}$ cm$^2$.

3. Computational and Theoretical Analysis

What are the circumstances under which it is possible the chronological measurements of cosmogenic isotopes? The main production of $^{53}$Mn and $^{10}$Be takes place due to the nuclear-active component of cosmic rays in the surface layer of rocks of a few hundred grams per cm$^2$ thick.

Let's assume that the surface layer of rocks is irradiated during a time $T$ needed for establishing equilibrium state on $^{53}$Mn (and hence on $^{10}$Be), that is $T \geq 10$ million years. Then the unit volume of rock will contain atoms $N_1$, atoms of $^{53}$Mn and $N_2$ atoms of $^{10}$Be in accordance with the relations

$$N_1 = N_{01} \sigma_1 \Phi_p / \lambda_1$$ \hspace{1cm} (2)

$$N_2 = N_{02} \sigma_2 \Phi_p / \lambda_2$$ \hspace{1cm} (3)

where $N_{01}$ –is the number of atoms $^{56}$Fe, $N_{02}$ –the total number of O, Si and Al atoms in the investigated volume, $\sigma_1$ and $\sigma_2$ – the averaged over spectrum production cross-sections for given chemical composition of $^{53}$Mn and $^{10}$Be respectively, $\Phi_p$ -the proton flux of CR, and $\lambda_1$, $\lambda_2$ -the decay constants for $^{53}$Mn and $^{10}$Be respectively.

Obviously, the equilibrium ratio $^{53}$Mn/$^{10}$Be is independent of the flow $\Phi_p$, i.e.
If then, for any reason, the surface layer will rapidly (in tens of thousands of years), shielded with sufficiently thick (for absorbing nuclear-active components of CR) layer of material (water, ice, etc.), the generation $^{53}$Mn $^{10}$Be practically stopped (except for the contribution of muons).

Since then, the ratio $N_1/N_2$ will be determined only by the decay of $^{53}$Mn and $^{10}$Be, i.e.

$$N_1/N_2 = \lambda_2 N_{01} \sigma_1 \exp(-T \lambda_1)/\lambda_2 N_{02} \sigma_2 \exp(-T \lambda_2).$$

This gives rise to the expression for the shielding time:

$$T = \ln \left( \frac{\lambda_1 \sigma_1 N_{01} / (\lambda_2 \sigma_2 N_{02})}{(\lambda_2 - \lambda_1)} \right).$$

Thus, determining the ratio of $^{53}$Mn/$^{10}$Be = $N_1/N_2$ in rock sample (and assuming known cross sections $\sigma_1$ and $\sigma_2$) we measure the absolute shielding time. In addition, from the expression (2) we can also determine the CR proton flux $\Phi_p$ at the time of exposure, i.e., before the shielding:

$$\Phi_p = \exp(T \lambda_1) N_{mes} \lambda_1/(N_{01} \sigma_1),$$

where $N_{mes}$ is the content of $^{53}$Mn in a sample of rock measured in a time $T$ after the shielding.

The above “shielding” has repeatedly occurred in the relatively recent (millions years ago) Earth’s geological history. This is also the fast enough (for the tens of thousands of years) immersing of land areas into the ocean as well as glaciations (Antarctica, Greenland). Since such events are stretched in time and occur at different locations, we are able to get a series of measurements of flow $\Phi_p$ in the time range between 0.1 to about 10 million years. Note again that the use of precisely these two (at least!) Isotopes $^{10}$Be and $^{53}$Mn allows determining both the time of shielding $T$ and the flux of cosmic rays $\Phi_p$ before shielding. According to geological estimates, the ice sheets of Antarctica and Greenland were formed only a few million years ago. Precise measurement of time is important.

Measuring the age of sedimentary layers requires separate consideration. Sedimentary layers are formed by the material for a long time lying on the surface and the material before entering the precipitate is irradiated with protons of cosmic rays at an unknown depth and chemical composition of the surrounding rocks, which defines the total flux and the energy of the protons in nuclear cascade in the ground. With high probability $^{10}$Be is in equilibrium with the proton flux (as it is produced in the reactions with O, Si, and Al elements, which collectively consist of 98% atomic concentration of rocks, and it simply has nowhere to go. In addition, the main part of $^{10}$Be is contained in sand silicate breed, migration from where is extremely difficult.

The situation with $^{53}$Mn is unclear. It is not known whether Fe enters the sedimentary layers have been completely cleared from $^{53}$Mn. Iron constitutes 5% by weight and less than 1% by atomic concentration of rock, i.e. there is a huge reservoir of solvent. Since iron dust particles before reaching the layer of sedimentary rocks are subjected to repeated recrystallization during the migration process, it is possible that in this case there is a complete separation of Fe and Mn. Where in this case leaves Mn - is unclear.

The preliminary Monte Carlo calculations of the accumulation rate of radioisotopes $^{10}$Be and $^{53}$Mn in the real ground (please see Fig. 2) showed that the measurement of the radioisotopes accumulation is really possible by means, for example, neutron activation analysis. We used the fluxes of cosmogenic protons and neutrons in accordance with paper [7], please see Figure 3. The angular distribution accepted in the form $f(\cos \theta) \propto (\cos \theta)^{1/3}$. 

**Figure 2.** Distribution of production rate of radioisotopes along depth in terrestrial ground calculated using a transport code SHIELD [6].

**Figure 3.** Fluxes of cosmogenic protons and neutrons at sea level [7].

### 4. Methods for Measurement of $^{53}$Mn Quantity

Methods for measurement of $^{53}$Mn quantity are well known. These include radiometric method, neutron - activation analysis, mass - spectrometric method, as well as recently intensively developing technique of accelerator mass - spectrometry, please see, for example, [8-9] and references therein.
Of all the above mentioned methods of isotope content determination the techniques of neutron activation analysis is relatively low-cost both in money and time. Therefore, it is likely this method will be used to study a large number of cores of terrestrial rocks from different regions. Neutron - activation method is quite acceptable, even in the case when a ballast ratio $^{55}$Mn/$^{54}$Mn is sufficiently low of about $10^{11}$, due to the fact that the ballast $^{58}$Mn has closed the so-called "semimagic" neutron shell of nuclei. Because of this background reactions $^{55}$Mn (n, 2n) $^{54}$Mn and $^{55}$Mn (γ, n) $^{54}$Mn during irradiation in the nuclear reactor are suppressed to a high degree due to the high threshold of these reactions. In addition, $^{54}$Mn has a very comfortable γ-line with the energy of 835 keV (100%). Therefore $^{53}$Mn measurement procedure can be simplified and made cheaper, which is very handy when a large number of measurements must be made. Moreover, in the case of neutron - activation analysis the tens and hundreds of samples can be simultaneously irradiated in large irradiation channels of the nuclear reactor. The measurements procedure can be sufficiently simplified due to using the precise flowing gas radiochemical method for detection of neutron flux in nuclear reactors [10, 11]. Measurement of the γ - lines on a semiconductor detector can be easily produced. Thus, we focus on the use of neutron - activation analysis as a more adequate to the task assigned. Of course $^{10}$Be concentration, will be determined by accelerator mass - spectrometry. But $^{10}$Be measurement technology is known for a long time and is well established. Regarding $^{53}$Mn with its massive nuclei, the measuring of its content with accelerator mass - spectrometry is considerably complicated, particularly due to the presence of background $^{53}$Cr.

5. Conclusion

The use of $^{53}$Mn content measurement techniques in iron-containing rocks can provide the valuable information about the geological and climatic processes on the Earth, as well as can evaluate the intensity of cosmic rays in the range of 0.1 - 10 million years.

Technological civilization in the world has only a few hundred years. Such a short period of observation of the universe does not allow getting any reliable data on the variations and cycles of cosmic radiation, which could occur at the origin and disappearance of powerful cosmic radiation sources such as supernovae, gamma-ray bursts and others. Therefore, any new information about the variations in the past of cosmic rays is extremely useful.

Such information could be extracted from the measurements, for example, with the lunar soil samples. The intensity of proton GCR on the lunar surface depending on the phase of the solar cycle is $2\sim4$ proton/cm$^2$sec, that is 3$\sim4$ order of magnitude higher than the nucleon flux of cosmogenic origin on the surface of the Earth (please compare Fig. 3). Fluxes of secondary nucleons in the lunar soil are maximized at a depth of about 1 m, and are greater than the flux of nucleons at the Earth surface up to a depth of 6 m or more [12-14]. Accounting for the GCR nuclei increases the flux of secondary nucleons, approximately 1.5 times.

Therefore, it will be much easier to study the production of cosmogenic $^{55}$Mn and $^{10}$Be and other isotopes in the lunar soil than in terrestrial samples. In this case it would be possible not only to monitor variations in the GCR for geological times, but, for example, date lunar craters. It is assumed sampling of relatively fresh craters with age of about 0.5-10 million years with a careful survey of soil condition. At such a relatively small interval of time the likelihood of a serious deep mixing is small. Micrometeorite erosion affects only a thin surface layer with thickness of 10-20 cm. In case of contact with the large crater of a meteorite large enough, this event can be easily identified.

Of course, the use of the lunar surface as a kind of "photographic plates" at the moment is only purely hypothetical. However, only the moon is the place where it is possible to get the most "pure" data on the variation of cosmic rays.

References

