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ORIGINAL ARTICLE

Early 18th century cosmic ray flux inferred from ^{44}Ti in Agen meteorite

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Abstract We report the measurement of radioactivity of cosmogenic ^{44}Ti in Agen meteorite, a H5 chondrite that fell in 1814. The ^{44}Ti activity in meteorites is related to centennial-scale changes in cosmic ray intensity caused by heliospheric magnetic field modulation in the interplanetary space between heliocentric distances of 1 and 3 AU. The measured low ^{44}Ti activity in Agen suggests a strong modulation of galactic cosmic rays at the turn of the 18th century, resulting in a low cosmic ray flux and is consistent with the linearly decreasing trend of GCR flux, modulated by the Gleissberg solar cycle during the past 250 years, as previously suggested by us.

Keywords Meteorites · Cosmogenic radionuclides · Solar activity—methods: gamma-ray spectrometry

1 Introduction

The intensity of galactic cosmic rays (GCR) is modulated by the heliospheric magnetic field as these energetic particles travel from the interstellar space to the inner solar system due to the heliospheric magnetic field. The energetic particles are scattered by magnetic irregularities in the heliosphere and are subject to processes such as convection and adiabatic deceleration in the expanding solar wind. Changes in heliospheric conditions due to the solar activity cause an overall variation in GCR intensities according to the time- and energy-dependent cosmic ray transport equation given by Parker (1965) leading to the well-known anticorrelation between sunspot number and cosmic ray flux. Several long-term proxies of solar activity (e.g., sunspots, geomagnetic indices, cosmogenic radionuclides and aurorae) suggest the presence of significant secular and periodic variations of the solar activity. This long-term variability has already produced a considerable impact on global climate in the past and may play a crucial role, together with the effects of human-induced environmental changes, in significant climatic excursions over the coming decades. It is therefore pivotal to investigate how these solar activity cycles have behaved in the past, both to understand the long-term behavior of the Sun and to forecast scenarios of future global climate change on Earth. Although the amplitudes and periodicities of the solar cycles are usually inferred from the different proxy indices of solar activity mentioned above, there is still some mismatch between the results obtained with different indices. One major factor is the influence of variations of the geomagnetic field which not only affect aurorae and several geomagnetic indices but also the production rates of cosmogenic nuclides resulting from GCR-induced spallation of Earth atmospheric constituents. The other major factor is the influence of climatic variations which alter the exchange and deposition processes in terrestrial reservoirs (e.g., ^{10}Be in ice sheets and ^{14}C in tree trunks) besides the imprecisely known redistribution mechanisms of these isotopes in the terrestrial archives (Usoskin et al. 2009). As for the forward models that rely only on the properties of active regions (sunspots, faculae) and the historical sunspot record to infer the Interplanetary Magnetic Field (IMF) near Earth (e.g., Solanki et al. 2000, 2002; Wang and Sheeley 2003),

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they all involve ad hoc factors that remain to be quantitatively established by independent means.

Cosmogenic isotopes produced in meteorites during their exposure to GCR in the interplanetary space are able to provide a direct measure of the GCR flux, without the complications produced by the terrestrial factors mentioned above (Taricco et al. 2006). Typical uncertainties in estimates from ^{10}Be and ^{14}C due to the geomagnetic shielding, climatic conditions and redistribution process are totally avoided in this case, since the nuclides are produced directly in the meteorite body in space. The activity of cosmogenic radioisotopes in meteorites corresponds to an integral of the balance between the isotopes production and decay, thus representing the time-integrated CR flux over a period determined by the mean life of the radioisotope so that it provides integrated mean fluxes rather than the more preferable differential values. By measuring the abundance of relatively short-lived cosmogenic isotopes in meteorites which fell at different times in the past, one can evaluate the variability of GCR flux. About 20 radioisotopes, such as ^{46}Sc, ^{22}Na, ^{14}C, ^{44}Ti, ^{39}Ar, and ^{26}Al, having mean life ranging from days to millions of years, have been measured in several stony meteorites. Cosmogenic radioisotopes are able to yield information about the intensity and variation of cosmic ray flux for a period of about 3 half-lives. Therefore it is possible to study GCR variations on different timescales by measuring radioisotopes having different half-lives (Bonino et al. 1995; Taricco et al. 2006, 2007).

A nearly ideal isotope for studying centennial-scale variability is the radioactive nuclide ^{44}Ti (half-life = 59.2 ± 0.6 years) that is produced by spallation reactions, mainly due to cosmic ray protons ($>$ 70 MeV) in meteoritic Fe and Ni in the body of a meteorite (Bonino et al. 1995). Because of its mean life, ^{44}Ti is relatively insensitive to variations in cosmic-ray flux on the 11-year cycle timescale but very sensitive to the average level of GCR flux and its variations on a centennial timescale. The ^{44}Ti activity in meteorites thus represents a powerful tool, allowing us to decouple past solar activity variations from terrestrial effects and providing an effective means for verifying models based on ^{10}Be and ^{14}C cosmogenic isotopes, as we have shown earlier (Usoskin et al. 2006). The radioactive decay chain ^{44}Ti/^{44}Sc/^{44}Ca leads to emission of γ-rays which can be detected with sensitive gamma ray spectrometers.

Taricco et al. (2006) measured the ^{44}Ti activity in 20 chondrites which fell during the period 1766 to 2001 and showed that the cosmic ray intensity between 1 and 3 AU, the orbital space of meteorites, has nearly linearly decreased at a rate of 18 % per century. Superimposed on this monotonous trend, a cyclic variation of 25 % with an apparent periodicity of 87 years, attributed to the Gleissberg cycle, was also inferred. The earliest trough of this cyclic variation in our measurements occurs around the year 1805. To confirm this very low activity, we have now analyzed Agen meteorite, which fell in 1814, closest to the minimum of ^{44}Ti activity. A preliminary measurement of Agen meteorite was performed earlier by our group (Taricco et al. 2006), but the detector available at that time did not allow us to achieve sufficient precision. For more accurate measurement of such a low activity, we have now set up an improved gamma ray spectrometer, composed by detectors with higher efficiency and resolution compared to the ones used earlier by our group on which most of the meteorites were measured. The new experimental set up (Colombetti et al. 2013) is described in Sect. 3.

Table 1 Characteristics of Agen meteorite and radiometric measurement

Class	H5
Date of fall	5 September 1814
Source of meteorite	Vatican Obs. Rome
Recov. mass	30 kg
Wt. of sample counted	683 g
Fe + Ni (%)	27.45 + 1.74
K (ppm)	785[a]
^{21}Ne Exp. Age	7 Ma
Counting time	362 days
^{44}Ti (C) cpd (1157 keV)	0.84 ± 0.14
^{40}K (N) cpm (1460 keV)	1.928 ± 0.003
Decay time	197 years
^{44}Ti activity[b]	1.10 ± 0.21 dpm/kg
Shielding factor (Torino = 1)	1.48
Corrected ^{44}Ti activity	5.57 ± 0.97 dpm/kg(Fe + Ni)

[a] Adopted average meteorite class values from Kallemeyn et al. (1989) and Mason (1971)

[b] Not corrected for shielding

2 The Agen meteorite

The Agen meteorite was seen to fall on Earth at noon of September 5, 1814 in Aquitaine, France. The recovered weight of the shower of stones was about 30 kg, the largest one weighing about 9 kg (Grady 2000). Agen has been classified as H type ordinary chondritic meteorite and belongs to the petrologic type 5. This H group chondrite has Fe abundance of about 27.45 % and Ni (1.74 %) by weight, which is especially favorable for ^{44}Ti production as compared to other ordinary chondrites. A fragment of this meteorite, weighing 683 g, was obtained from the Vatican Observatory museum of Rome and has been made available to us for gamma-ray measurements at the Research Station of Monte dei Cappuccini (INAF-OATo, Italy).

3 Experimental setup

By virtue of the low production cross sections from Fe, and especially because of the decay of the activity since the fall of the meteorite, the ^{44}Ti activity in meteorites is extremely low for ancient meteorites. The measurement of such low levels of radioactivity in meteorite samples thus requires highly sensitive detectors and special configuration. Furthermore, stray radiation from the environmental rocks and penetrating cosmic rays create much interference with the signal and so that both high specificity and selectivity are required to minimize the background. To achieve a good performance, additionally, there are further constraints since meteorites are precious extraterrestrial samples and can not be destroyed or dissolved to concentrate the radioactive elements. We therefore developed a spectrometer that is capable of non-destructive measurements of meteorite samples up to 1 kg mass and a specific data acquisition system tailored to the decay scheme of ^{44}Ti and its decay chain. ^{44}Ti decays to its radioactive daughter, ^{44}Sc ($T_{1|2} = 3.93$ h), emitting gamma rays of 68 and 78 keV. ^{44}Sc subsequently decays with a 3.9 hour half-life to ^{44}Ca, emitting a positron in coincidence with a 1157 keV γ-ray. Our gamma ray spectrometer, designed according to the decay scheme of ^{44}Sc, allows a highly sensitive and selective measurement of 1157 keV γ-ray, by detection of one (511 keV) or two (1022 keV) annihilation photons in coincidence. The 3 kg High-Purity Germanium (HPGe) detector, used as the main detector, has a relative efficiency of 147 % (wrt the standard 7.5 cm NaI scintillator, used as a reference), a resolution of 1.85 keV and a peak-to-Compton ratio of 104 for the 1332.5 keV ^{60}Co gamma-rays. It is surrounded by a Thallium-doped Sodium Iodide (NaI(Tl)) annular single crystal and a NaI(Tl) cylindrical plug at the top (total mass 90 kg), as an anticoincidence umbrella for rejecting the stray ambient radiation and penetrating cosmic radiation. The scintillator is coupled to 7 photomultipliers for better optical efficiency. The assembly is housed in a sequential Pb-Cd-Cu-Polyethylene shield to absorb and eliminate environmental gamma rays. The outermost shield is made of 20 cm thick high-purity lead, internally lined with 1 mm of Cd, 5 cm oxygen-free high thermal conductivity (OFHC) copper and the empty space surrounding the detector is filled with polyethylene to reduce the amount of ambient radon present in the air. To shield off penetrating cosmic rays, the spectrometer is located underground (under 70 meter water equivalent overburden of soil) in the Research Station of Monte dei Cappuccini in Torino, Italy. ^{44}Sc(^{44}Ti) can thus be efficiently counted, mainly because the interference due to the environmental 1155 keV gamma ray from the ambient ^{214}Bi, a daughter product of radon (uranium), is minimized. A detailed description of this system is given in Taricco et al. (2007) and Colombetti et al. (2013).

4 Results and discussions

In Fig. 1 we show the spectra of Agen meteorite, obtained (a) in normal mode (HPGe alone, black line) and (b) in coincidence mode (HPGe in coincidence with NaI, blue line). Besides the cosmogenic ^{26}Al and ^{44}Ti, the other peaks are due to the background of naturally occurring potassium, uranium and thorium nuclides and their gamma-emitting decay products. ^{214}Bi comes from ^{238}U decay chain, while ^{208}Tl and ^{228}Ac from ^{232}Th chain. The 511 keV peak is from positron annihilations. The main source of background in the immediate adjacent bins towards lower energy of ^{44}Ti peak (1157 keV) is that due to the environmental ^{214}Bi at 1155.19 keV, which is omnipresent. This peak is partially superimposed on the ^{44}Ti peak, and although unimportant for fresh meteorite falls where ^{44}Ti activity is high, becomes increasingly important for older falls (in which ^{44}Ti has significantly decayed), as in the case of Agen meteorite. Thanks

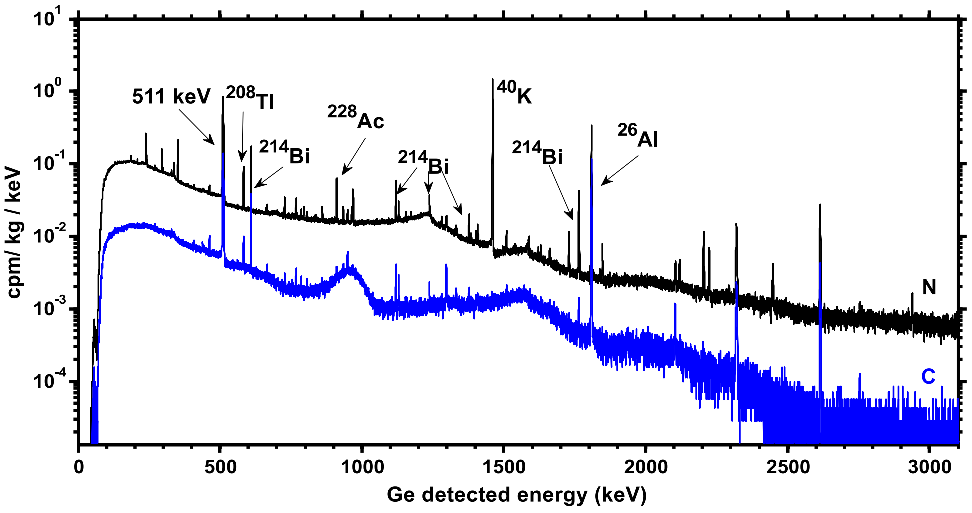

Fig. 1 γ-ray spectrum of the Agen meteorite fragment in *normal mode* (HPGe alone) (*black line*) and in *coincidence mode* (HPGe in coincidence with NaI) (*blue line*)

to the coincidence configuration described above, the ^{214}Bi peak is strongly reduced and the Compton level reduced by a factor of about 20, without much loss in the counting efficiency.

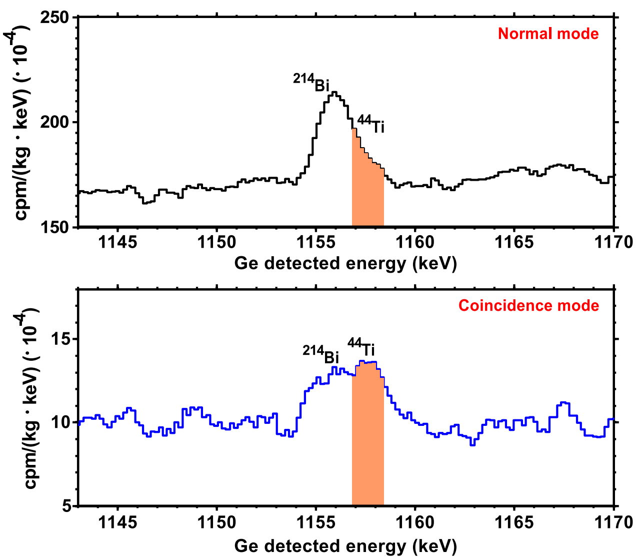

Fig. 2 Detail of the spectrum shown in Fig. 2. The spectrum in *coincidence mode* (HPGe in coincidence with NaI) (*blue line*) shows that the background is considerably reduced and the ^{44}Ti peak becomes relatively prominent

The enlarged ^{44}Ti spectral region is shown in Fig. 2 revealing that the interference due to ^{214}Bi, which is present in normal mode, is significantly reduced in coincidence mode, whereas the ^{44}Ti peak becomes relatively prominent. The ^{44}Ti count per day deduced from the coincidence spectrum (reported in Table 1) has to be corrected for detection efficiency, Fe and Ni abundance and decay of the radioisotope since the fall of the meteorite. Moreover, in order to relate the measured ^{44}Ti activity to cosmic ray modulation, the shielding effect of the fragment in space has to be taken into account, since the production is meteorite-size and fragment-depth dependent. Here we follow the procedure of Bhandari et al. (1980) using ^{26}Al activity, compared to its production rate, as proxy for shielding, normalizing to Torino meteorite as the shielding reference. After applying these corrections, we have obtained the ^{44}Ti activity at the time of fall (5.57 ± 0.97 dpm/kg(Fe + Ni)). This value is shown in Fig. 3, together with the measurements of the other meteorites covering the period 1766 to 2001, previously analyzed by our group. The error bars for all meteorites shown in Fig. 3 represent the statistical uncertainties of measurements (1σ). Other sources of error are due to errors in the concentration of K, Fe and Ni and in the shielding depth evaluation. These corrections are usually small ($< 5\ \%$) and do not affect the results reported here.

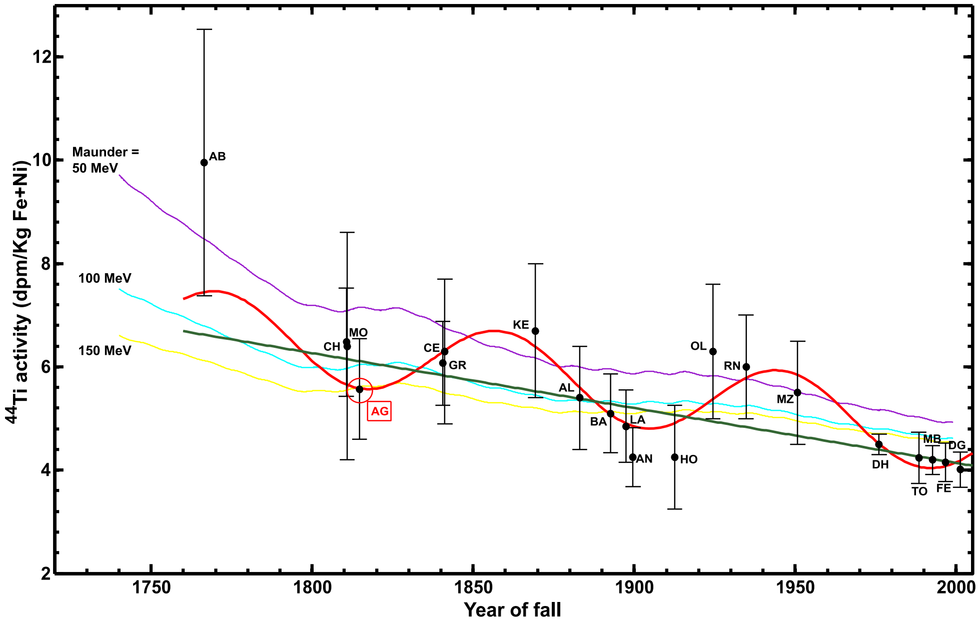

Fig. 3 Measured ^{44}Ti activity (dpm/kg Fe + Ni), corrected for shielding and target element composition as a function of time of fall of chondrites covering the period 1766 to 2001 AD. Superimposed to the data, (a) the best fit curves using a straight line (*green curve*), (b) a sinusoid plus linear trend $y(t) = a + bt + \sin(d + 2\pi T/t)$ (*red curve*) and (c) activities calculated using the production model of Michel and Neumann (1998) corresponding to different assumptions of the modulation parameter Φ during the Maunder minimum (*violet*, *cyan* and *yellow curves*; see Taricco et al. 2006 for details) are shown. Labels in the figure correspond to the measured meteorites: Albareto (AB), Charsonville (CH), Mooresfort (MO), Agen (AG), Cereseto (CE), Gruneberg (GR), Kernouve (KE), Alfianello (AL), Bath (BA), Lancon (LA), Allegan (AN), Holbrook (HO), Olivenza (OL), Rio Negro (RN), Monze (MZ), Dhajala (DH), Torino (TO), Mbale (MB), Fermo (FE) and Dergaon (DG).

The low ^{44}Ti activity of Agen meteorite relates to a minimum phase of the cosmic ray flux a few decades before the fall (1814), since the activity of a cosmogenic isotope represents an integral of the balance between the isotope's production and its decay over a few half lives.

The Agen measurement confirms that the ^{44}Ti activity decreased over 235 years (Taricco et al. 2006) by 2.6 dpm/kg(Fe + Ni) corresponding to about 40 % since 1766. Superposed on this nearly linear trend, we find a centennial modulation of GCR due to the Gleissberg solar cycle of period T (87.3 ± 8.7) years. This value has been obtained using a sinusoidal plus linear trend fit (red line in Fig. 3), taking the measurement uncertainties into account. This fit yields a higher correlation coefficient ($R^2 = 0.84$ with root mean square error RMSE = 0.56) as compared to the one obtained using only a straight line (green line in Fig. 3, for which $R^2 = 0.65$ and RMSE = 0.76).

In Fig. 3, the comparison of the observed ^{44}Ti activity in meteorites with the activities calculated assuming different values for the modulation parameter during the Maunder minimum and the production model of Michel and Neumann (1998) is also shown (see Taricco et al. 2006 for details of the model). We notice a general agreement between the measured and the estimated decreasing trend of the activity over the past 235 years. The centennial variation in the measured series seems to be larger than indicated by the calculated profiles. This suggests that during prolonged periods of low solar activity, such as the Gleissberg minima, the cosmic ray flux was significantly higher than the values computed on the basis of the relationship of solar activity and GCR flux (Taricco et al. 2006).

5 Conclusions

Using the spectrometer developed by our group at the Laboratory of Monte dei Cappuccini in Torino, we measured the ^{44}Ti activity in Agen meteorite, which reveals the mid 18th century minimum of GCR flux, related to the Gleissberg solar cycle. This value confirms the decreasing trend of the GCR flux in the interplanetary space (1 to 3 AU) during the past 235 years, as previously suggested (Taricco et al. 2006). The decrease of about 40 % is consistent with the calculated GCR flux in the past based on the evolution of the Sun large-scale open magnetic field (Solanki et al. 2000). The activity of Agen and of the other chondrites which fell during the past three centuries is higher compared to that of the meteorites which fell after 1970, revealing that the GCR flux during the XVIII century was higher than during the last few decades.

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