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Radionuclide activities in recent chondrite falls determined by gamma-ray spectrometry: Implications for terrestrial age estimates

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(Received 16 August 2018; revision accepted 31 October 2019)

Abstract–Radionuclide activities were measured in the low-background gamma-ray spectrometry facility GeMSE in eight meteorite falls (Lost City, Tamdakht, Huaxi, Boumdeid, Xining, Kamargaon, Degtevo, and Ouidiyat Sbaa) and two finds (SaU 606 and Mürtchenstock) to evaluate the use of radionuclides for terrestrial age estimates. Results indicate that these meteorites were all derived from small- ($r < 25$ cm) to medium-sized ($r < 65$ cm) meteoroids. Short-lived ^{48}V ($t_{1/2} = 16.0$ d) and ^{51}Cr ($t_{1/2} = 27.7$ d) were only detected in Ouidiyat Sbaa (EH), while ^7Be ($t_{1/2} = 53.1$ d) was also detected in Degtevo (H) and Kamargaon (L), in agreement with reported fall dates. The $^{22}\text{Na}/^{26}\text{Al}$ activity ratio in Huaxi agrees with the previously reported short cosmic-ray exposure age of this meteorite while $^{22}\text{Na}/^{26}\text{Al}$ in Kamargaon likely records a complex exposure history. Bayesian statistical analysis verifies the detection of very low activities of ^{44}Ti ($t_{1/2} = 60$ a) in the relatively large H chondrites (>100 g) Degtevo, Huaxi, Tamdakht, Lost City, and SaU 606. Additionally, large samples from Ouidiyat Sbaa (EH) and Kamargaon (L) gave positive detections. For H chondrite target compositions, detected $^{44}\text{Ti}(\text{Fe}+\text{Ni})/^{26}\text{Al}$ averaged 0.055 ± 0.013 . Activities of ^{22}Na and ^{54}Mn in SaU 606 show that this meteorite fell between July and September 2012, making SaU 606 the second recent fall from Oman identified using gamma-ray spectrometry. The upper activity limit of ^{22}Na in the Mürtchenstock meteorite shows that it fell prior to year 1999 and is not related to a bolide observation in 2015. Mürtchenstock shows $^{137}\text{Cs} \sim 10\times$ higher than previously determined in Oman meteorites, likely due to Chernobyl fallout.

INTRODUCTION

Many cosmogenic radionuclides are produced in meteoroids due to irradiation by cosmic rays (e.g., Leya and Masarik 2009). Measurements of radionuclides can provide important information about the irradiation history of meteorites such as the pre-atmospheric shielding parameters and exposure times (e.g., Li et al. 2017). The radionuclide record preserved in observed meteorite falls and meteorite finds can also be used to verify fall observations and provide accurate terrestrial age determinations. Since early 2016, the low-background Germanium Material and Meteorite Screening Experiment (GeMSE), (von Sivers et al. 2016)

situated in a tunnel below Vue-des-Alpes in the Swiss Jura mountains, has dedicated substantial screening time to meteorite fall samples. The goals of the campaign are a set of cosmogenic radionuclide measurements which can be used to evaluate measurements used to screen Oman finds and at the same time explore methods to increase measurement precision and improve measurement routines in ongoing and future projects. The robustness of future measurement evaluations will be improved by using the same methods on all samples. As an analytical method, gamma-ray spectrometry has the key advantages that the bulk activities of gamma-emitting isotopes can be obtained simultaneously from one single, non-destructive measurement. In the GeMSE

facility, high measurement efficiencies, very competitive background levels, and a large sample cavity enable accurate and precise measurements of gamma rays, when analyzing sample masses ranging from tens of grams to several kilograms. On a short time scale, weeks to months after a meteorite fall, short-lived cosmogenic isotopes (e.g., ^{48}V , $t_{1/2} = 16$ d) can be used to estimate the time a meteorite fell with a precision that largely depends on the time duration between the fall and start of measurement. On an intermediate time scale, ^{22}Na ($t_{1/2} = 2.60$ a) and, when available with sufficient activities, ^{60}Co ($t_{1/2} = 5.27$ a) can be used to identify recently fallen finds. For the purpose of identifying samples with slightly older terrestrial ages, ^{44}Ti ($t_{1/2} = 60.0$ a) is the only available gamma-emitting cosmogenic isotope which has previously been detected in meteorites. The ^{44}Ti activity in meteorites has been suggested to record decade- to century-scale variations in the cosmic irradiation history (Bonino et al. 1995; Taricco et al. 2006) and can potentially be used to determine the terrestrial age of samples which fell several decades or even centuries ago. Low activities and partly overlapping gamma lines make this isotope especially hard to measure in meteorites. Discrepancies between calculated production rates of ^{44}Ti (Michel and Neumann 1998) and detected activities in meteorites (e.g., Taricco et al. 2006) make it difficult to estimate minimum terrestrial ages based on the upper activity limits from measurements where ^{44}Ti was not detected.

This study presents an internally consistent set of cosmogenic and other radionuclide activities measured in samples from eight different chondrites with known dates of fall. We evaluate the results in the context of analytical precision and accuracy of bulk sample measurements as well as cosmogenic radionuclide production rates. We further present the results of measurements performed on two samples from meteorite finds with unknown fall dates and discuss the implication of these in the light of our data from the fall sample measurements. With this study, we have several goals: (1) test the ability to measure ^{44}Ti and quantify the activity of this isotope in recent meteorite falls with the potential aim of using ^{44}Ti for the recognition of decade- to century-old falls; (2) determine the activities of cosmogenic radioisotopes in a series of recent meteorite falls and correlate these to the respective irradiation history and meteoroid size; (3) use radioisotope data for the verification of reported fall dates and, in case of finds, for the determination of terrestrial ages; (4) obtain important information regarding the potential of finding additional meteorites from the same fall through estimates of pre-atmospheric radii based on activities of ^{26}Al and ^{60}Co .

Meteorite Samples

Gamma-ray spectrometry measurements in GeMSE were performed on samples from eight different meteorite falls and two finds (Table 1). In addition to four meteorites that were not analyzed previously by gamma-ray spectrometry, the fall samples include Lost City, Tamdakht, Boumdeid (2011), and Kamargaon. Radionuclide data from four different fragments of the Lost City meteorite are included in a large compilation of measurement data from meteorites that fell between 1967 and 1977 (Evans et al. 1982). Gamma-ray spectrometry and noble gas data on the Tamdakht meteorite were presented in a previous study encompassing four different meteorite falls (Alexeev et al. 2012). Boumdeid (2011) (Buhl et al. 2014) and Kamargaon (Ray et al. 2017) have been described in previous detailed petrographic studies which included radionuclide measurements. Two fragments each from the recent Chinese falls Huaxi and Xining (designated Huaxi A, B and Xining A, B) were analyzed. The Huaxi samples appear to have come from one single stone which made up the initial main mass. This stone shattered already upon impact according to a recent detailed petrographic study which included noble gas measurements (Li et al. 2017). The Xining samples have less well-documented find conditions as the initial find consisted of 10 individual stones, scattered across a $\sim 25 \times 5$ km area, which were first distributed to private collectors. Degtevo is the second most recent (2016) recovered meteorite fall in Russia which was classified as an H5 ordinary chondrite as part of this study. Oudiyat Sbaa fell in 2016, constituting the latest observed enstatite chondrite fall and one out of in total 10 EH chondrite falls known to this date. In addition to the eight falls, two different meteorite finds were selected for the current study: Sayh al Uhaymir 606 (SaU 606) is an ordinary chondrite of type H5 which was found 2017 during a search campaign in Oman organized by the Omani-Swiss meteorite search project. The samples recovered during this project are stored at the Natural History Museum Bern as parts of the Omani-Swiss meteorite search collection. The SaU 606 meteorite was immediately selected for further study based on a very low degree of terrestrial weathering (W1), which could potentially indicate that it fell in recent years. Additionally, one recent Swiss find was analyzed. Mürtschenstock is an L6 type ordinary chondrite which was found during 2017 in the Canton of Glarus, central Swiss Alps. Due to its relatively low degree of weathering (W1) and poor constraints on weathering rates of chondrites in humid climates, it was initially suggested that this meteorite might be connected to a bolide observation in

Table 1. Details about studied falls and samples. Screening times and determined bulk densities are listed. The fall dates and values for recovered mass were taken from the *Meteoritical Bulletin* database, accessed January–May 2019. For Huaxi and Xining, A and B denote subsamples from the respective meteorites.

Meteorite samples						
Falls	Recovered mass (kg)	Fall date	Source, Inv. Nr.	Sample mass (g)	Screening time (days)	Measured density (g cm ⁻³)
Ouidiyat Sbaa (EH5)	23.8	November 18, 2016	Loan from Abdelaziz Habibi/ Tony Irving, University of Washington	191	9	3.62
Huaxi; A, B (H5)	1.6	July 13, 2010	Loan from Shijie Li, Chinese Academy of Sciences, Guiyang	163, 117	19, 22	3.17, 3.13
Xining; A, B (L5)	100	February 11, 2012	Loan from Shijie Li, Chinese Academy of Sciences, Guiyang	38, 30	14, 12	3.44, 3.37
Tamdakht (H5)	100	December 20, 2008	NMBE 42971	128	12	3.57
Boumdeid (2011), (L6)	3.6	September 14, 2011	NMBE 41388	51	21	3.38
Degtevo (H5)	2.1	March 20, 2016	Intact main mass loaned from Marc Jost, Swiss Space Jewels, Brügg, Switzerland	2090	15	n.d.
Kamargaon (L6)	12.1	November 13, 2015	Loan from Dipankar Pathak, University of Bern/Aditi Bezbaruah, Jorhat Engineering College	176	14	n.d.
Lost City (H5)	17	January 4, 1970	Loan from National Museum of Natural History (Smithsonian), Washington, DC, USNM 4848	359	21	3.45
Finds	Recovered mass (kg)	Found		Sample mass(g)		
SaU 606 (H5)	0.137	Oman 2017	Omani-Swiss meteorite collection at NMBE (1702_0369)	137	15	3.43
Mürtschenstock (L6)	0.355	Switzerland 2017	Main mass on loan from ETH Zurich/ Naturwissenschaftliche Sammlungen Glarus/Andreas Stucki (complete individual)	355	8	3.27

n.d. = not determined.

southern Germany-central Switzerland on March 15, 2015.

METHODS

Gamma-Ray Measurements

Gamma activities were measured in the GeMSE facility during 2016–2017 using an ultra-low background high-purity germanium detector from Canberra Industries. Total measurement times were 1–3 weeks for each individual sample. The GeMSE facility and the general analytical procedures are described in

detail in von Sivers et al. (2016). All measured radionuclides, along with the corresponding half-lives and gamma lines, are listed in Table 2. Titanium-44 ($t_{1/2} = 60.0$ a) was analyzed via its short-lived daughter isotope ^{44}Sc ($t_{1/2} = 3.97$ h).

Activity Calculations

Counting efficiencies were simulated using GEANT4 v9.6p03 (Geometry And Tracking, Agostinelli et al. 2003) for the GeMSE setup and representations of the individual sample geometries. With carefully measured sample geometries (including volume and density), this

Table 2. Analyzed isotopes with half-lives and gamma line energies.

Isotopes	Half-lives ($t_{1/2}$)	Gamma lines (keV)
^{26}Al	717 ka	1808.7
$^{44}\text{Ti} (^{44}\text{Sc})$	60 a	1157.0
^{60}Co	5.27 a	1173.2, 1332.5
^{22}Na	2.60 a	1274.5
^{54}Mn	312.3 d	834.8
^{57}Co	271.8 d	122.1, 136.5
^{46}Sc	83.79 d	889.3, 1120.5
^{56}Co	77.27 d	846.8, 1238
^{58}Co	70.86 d	810.8
^7Be	53.12 d	477.6
^{51}Cr	27.70 d	320.1
^{48}V	15.97 d	983.5, 1312.1
^{40}K	1.248 Ga	1460.8
^{137}Cs	30.08 a	661.7

procedure reproduces detection efficiencies, in the relevant energy range, with accuracies better than 2% as determined by previous measurements with calibrated samples (von Sivers et al. 2016). Average bulk densities of the respective chondrite groups (Consolmagno et al. 2008) were generally used as input when running the simulations. Additionally, we used an optical 3-D scanner to determine the volume of most analyzed samples so that the bulk density input values could be adjusted for samples that were found to deviate significantly from the average group density. Homogeneous elemental compositions of ordinary chondrite and enstatite chondrite groups (Wasson and Kallemeyn 1988) corresponding to the analyzed samples were assumed representative and implemented in the simulations. Sample geometries were represented as cuboids, ellipsoids, or cut ellipsoids. This simplification of natural meteorite sample geometries adds extra uncertainties (~5%) to the simulated detection efficiencies which were accounted for through expanded systematic uncertainties. In the future, we plan to directly import the scanned 3-D sample geometries into Geant4. Ratios of suitable cosmogenic isotopes ($^{22}\text{Na}/^{26}\text{Al}$, $^{44}\text{Ti}/^{26}\text{Al}$), emitting gamma rays at similar energies, were calculated separately. The gamma-ray efficiency ratios of such pairs vary substantially less between different sample geometries compared to the detection efficiencies for single gamma lines at the corresponding energies. The measurement uncertainties were determined by comparing results from multiple simulations with densities, compositions, and masses that cover all studied physical samples and different placements in the sample cavity. In this range, the average ratio of simulated detection efficiencies ($\epsilon_{\text{N}}^{\text{a}}/\epsilon_{\text{N}}^{\text{b}}$) in GeMSE has an uncertainty below 1.5% 2RSD for $\epsilon^{22}\text{Na}/\epsilon^{26}\text{Al}$. This uncertainty also applies

for $\epsilon^{44}\text{Ti} (^{44}\text{Sc})/\epsilon^{26}\text{Al}$ but within slightly smaller intervals of sample mass. When calculating the activities of samples placed directly on the detector endcap, we implemented expanded uncertainties of 5% after carefully recording the placing and dimensions of each sample. The main mass of Degtevo could only fit next to the endcap and a more conservative value of 10% was used. Radioactive decay constants, gamma-ray intensities, and energies were all in accordance with the National Nuclear Data Center's online archives (Kinsey 1997), accessed 2017–2018. The determination of isotope activities or upper limits in the samples is based on Bayesian statistics (see von Sivers et al. 2016 for details). The significance of a signal is evaluated using the so-called Bayes Factor (BF). In meteorites with known fall dates or for which a young terrestrial age is otherwise indicated, we can assume that some amount of ^{44}Ti is present, based on cross section data and previously published gamma-spectrometry data. In this study, we choose to present and evaluate the calculated activities of ^{44}Ti and other more prominent short-lived cosmogenic isotopes for analyses yielding a Bayesian statistical probability of detection defined as “positive evidence, $\text{BF} \geq 3$ ” or stronger (Kass and Raftery 1995).

RESULTS

Bulk Densities

Bulk densities determined through optical 3-D scanning range between 3.1 and 3.6 g cm⁻³ (Table 1). The largest variability occurs among the studied H chondrite fragments. The two fragments from the Huaxi main mass have the lowest measured bulk densities, 3.13–3.17 g cm⁻³, of all analyzed samples. In contrast, at 3.57 g cm⁻³, the Tamdakht meteorite sample is only slightly less dense than Oudiyat Sbaa EH chondrite fragment, which has a bulk density of 3.62 g cm⁻³. All L chondrite samples had bulk densities of 3.27–3.42 g cm⁻³. Shape volumes were not obtained from the Kamargaon and Degtevo samples since these samples were no longer available when we had gained access to the optical scanner.

Radionuclide Activities

Results from the gamma-spectrometry measurements are listed in Table 3, where radionuclide activities in meteorites with known fall dates have been corrected to the time of fall. Separately calculated activity ratios are given at the end of the table together with the BFs for positive detections of ^{44}Sc , the daughter isotope of ^{44}Ti . We detected ^{26}Al in all samples at activities of ~35

Table 3. Calculated activities and activity ratios in samples from meteorite falls (dpm per kg at date of fall) and finds (dpm per kg at finished measurement)

Isotopes	Falls					Finds						
	OudiyatSbaa EH	Degtevo H	Kamargaon L	Xining A L	Xining B L	Boumdeid L	Huaxi A H	Huaxi B H	Tamdakht H	Lost city H	SaU 606 H	Mürtschenstock L
²⁶ Al	54.9 ^{+3.6} _{-3.3}	48.5 ^{+3.7} _{-1.8}	69.5 ^{+4.0} _{-3.4}	59.4 ^{+3.7} _{-3.4}	57.5 ^{+4.2} _{-3.2}	50.8 ^{+3.4} _{-2.3}	33.7 ^{+2.1} _{-0.6}	35.2 ^{+2.2} _{-1.7}	56.7 ^{+3.8} _{-4.3}	49.0 ^{+2.9} _{-2.2}	38.6 ^{+2.1} _{-2.2}	50.5 ^{+3.1} _{-2.5}
⁴⁴ Ti/ ⁴⁴ Sc	1.0 ^{+0.3} _{-0.3}	0.8 ^{+0.1} _{-0.1}	0.7 ^{+0.3} _{-0.4}	<1.6	<1.7	<0.8	0.6 ^{+0.3} _{-0.2}	0.5 ^{+0.3} _{-0.2}	1.3 ^{+0.3} _{-0.4}	0.6 ^{+0.3} _{-0.3}	0.7 ^{+0.3} _{-0.3}	<0.9
⁶⁰ Co	<0.43	0.3 ^{+0.1} _{-0.1}	<0.5	2.2 ^{+0.7} _{-0.6}	1.4 ^{+0.6} _{-0.6}	<1.0	<0.7	<0.6	33.4 ^{+1.4} _{-1.6}	<76.5	<0.5	<0.2
²² Na	61.3 ^{+3.7} _{-2.9}	66.0 ^{+3.2} _{-3.3}	61.2 ^{+4.3} _{-2.4}	95.3 ^{+7.4} _{-6.4}	102.6 ^{+8.2} _{-7.7}	84.4 ^{+5.8} _{-4.7}	76.8 ^{+4.1} _{-4.7}	77.8 ^{+5.5} _{-4.1}	111.2 ^{+7.9} _{-6.9}	n.d.	18.4 ^{+1.1} _{-1.2}	<0.4
⁵⁴ Mn	68.5 ^{+4.1} _{-3.3}	56.7 ^{+3.0} _{-2.1}	41.5 ^{+2.3} _{-2.2}	73.6 ^{+20.0} _{-17.0}	119.0 ^{+19.5} _{-25.7}	68.3 ^{+4.2} ₋₇	80.5 ^{+15.2} _{-18.4}	67.8 ^{+23.1} _{-17.8}	n.d.	n.d.	1.1 ^{+0.2} _{-0.2}	n.d.
⁵⁷ Co	8.0 ^{+0.6} _{-0.5}	7.5 ^{+0.7} _{-1.3}	4.2 ^{+0.5} _{-0.3}	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
⁴⁶ Sc	9.0 ^{+0.8} _{-0.7}	10.4 ^{+0.9} _{-0.7}	3.6 ^{+1.3} _{-0.3}	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
⁵⁶ Co	7.1 ^{+0.7} _{-0.7}	7.1 ^{+0.8} _{-0.7}	5.5 ^{+1.3} _{-1.1}	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
⁵⁸ Co	6.0 ^{+0.7} _{-0.4}	6.5 ^{+1.0} _{-1.1}	2.4 ^{+1.1} _{-1.0}	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
⁷ Be	54.2 ^{+5.8} _{-5.7}	58.7 ^{+15.0} _{-15.4}	51.3 ^{+16.3} _{-20.5}	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
⁵¹ Cr	54.4 ^{+14.4} _{-11.2}	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
⁴⁸ V	46.9 ^{+7.7} _{-5.4}	* n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
⁴⁰ K	991 ⁺⁶⁴ ₋₄₂	1331 ⁺¹¹⁷ ₋₁₆₄	1456 ⁺⁸² ₋₆₆	1407 ⁺⁷⁴ ₋₇₁	1230 ⁺⁸⁰ ₋₅₂	1453 ⁺⁸⁸ ₋₆₄	1242 ⁺⁶⁰ ₋₅₈	1239 ⁺⁷⁹ ₋₄₇	1151 ⁺⁷² ₋₄₆	1306 ⁺⁶³ ₋₅₁	1257 ⁺⁶⁹ ₋₅₉	1144 ⁺⁶⁸ ₋₄₉
¹³⁷ Cs	<0.3	<0.03	<0.4	<0.3	<0.5	<0.2	<0.1	<0.2	<0.29	<0.7	<0.5	42.2 ^{+3.1} _{-1.4}
²² Na/ ²⁶ Al	1.10 ^{+0.05} _{-0.06}	1.35 ^{+0.05} _{-0.05}	0.89 ^{+0.04} _{-0.04}	1.60 ^{+0.15} _{-0.14}	1.76 ^{+0.18} _{-0.19}	1.65 ^{+0.10} _{-0.12}	2.27 ^{+0.14} _{-0.12}	2.22 ^{+0.14} _{-0.15}	1.93 ^{+0.15} _{-0.13}	n.d.	0.48 ^{+0.03} _{-0.03}	<7.9 × 10 ⁻³
(⁴⁴ Ti/ ²⁶ Al) × 10 ⁻²	1.9 ^{+0.5} _{-0.6}	1.8 ^{+0.2} _{-0.4}	0.9 ^{+0.5} _{-0.4}	<2.7	<2.9	<1.7	1.7 ^{+0.7} _{-0.5}	1.6 ^{+0.8} _{-0.7}	2.3 ^{+0.6} _{-0.7}	1.2 ^{+0.6} _{-0.6}	1.8 ^{+0.7} _{-0.9}	<1.7
BF (⁴⁴ Sc)	50	9 × 10 ⁶	3	4 × 10 ⁻¹	3 × 10 ⁻¹	5 × 10 ⁻¹	30	5	3 × 10 ²	3	6	1

Further sample details are given in Table 1. Uncertainties of the detection efficiencies were 10% for Degtevo, for other samples 5%, and for activity ratios 1.5%. Maximum activity limits are given at a 95% credible interval and detected activities at 68%. The Bayes Factor (BF) corresponding to positive detection of ⁴⁴Sc is given at the end of the table. Values for ⁴⁴Ti/²⁶Al are listed in centiles. Dpm per kg for ⁴⁰K corresponds to 1.86 × concentration (ppm).

*Detection efficiencies and upper activity limits at the time of fall were not determined (hence n.d.) for short-lived isotopes which were inaccessible due to the time duration between fall and analysis (generally ~5 to 8 half-lives depending on saturation activities in chondrites).

to 70 dpm per kg. Cobalt-60 was detected in Tamdakht, Xining, and Degtevo, where the lowest activity, 0.3 dpm per kg, was observed when analyzing the Degtevo main mass. In both fragments of the Huaxi meteorite, ^{54}Mn was detected at a time equivalent of seven to eight half-lives after the fall. Isotopes with half-lives shorter than 300 days were detected in Kamargaon, Degtevo, and Oudiyat Sbaa. $^{137}\text{Cesium}$ was not detected in any of the meteorite fall samples, it was also not detected in SaU 606. The Mürtschenstock spectrum showed a prominent peak in the energy region corresponding to its presence. The activities of ^{40}K were ~ 900 to 1500 dpm per kg, in agreement with the compositional range (~ 400 to 1000 ppm) observed in ordinary and enstatite chondrites (Kimura et al. 2018). SaU 606 had clearly detectable activities of ^{22}Na and ^{54}Mn . In the Mürtschenstock sample, ^{26}Al was the only cosmogenic isotope that could still be detected. Due to higher concentrations of the target elements Fe and Ni, ^{44}Ti is expected to be easier to detect in H and EH chondrites than in L chondrites. We were able to detect ^{44}Ti in only one L chondrite sample, Kamargaon. Positive evidence for detection of ^{44}Ti was obtained (BF=3) for the Kamargaon and Lost City samples while even higher BF's were obtained for other H chondrite samples and the Oudiyat Sbaa (EH) fragment. Together, this indicates that the strength of the statistical evidence for positive detection of ^{44}Ti in meteorite falls, apart from the terrestrial age of a sample, shows an expected correlation with the sampled mass and concentration of target elements (Fe+Ni). Detected activities of ^{44}Ti correspond to less than 1.0 dpm per kg in all samples except the Oudiyat Sbaa and Tamdakht fragments. The generally high uncertainties (up to 50%) of the calculated $^{44}\text{Ti}/^{26}\text{Al}$ activity ratios ($0.9\text{--}2.3 \times 10^{-2}$) are due to the low activities of ^{44}Ti and resulting low count rates from ^{44}Sc . The activity ratios of $^{22}\text{Na}/^{26}\text{Al}$ were 1.1–1.9 in all fall samples except those from the Huaxi and Kamargaon meteorites which gave high and low ratios of ~ 2.2 to 2.3 and ~ 0.9 , respectively.

DISCUSSION

Bulk Densities

3-D scanning techniques have gained increased attention for their potential in noninvasive bulk density determinations of meteorite samples (e.g., Fry et al. 2018). For the purpose of precisely simulating gamma-ray detection efficiencies, this method is especially suitable since a samples volume and shape can be accurately recorded before these and other characteristics are implemented in the simulation code.

Fig. 1. Bulk densities obtained with an optical 3-D scanner and the corresponding sample masses. Common bulk densities for L, H, and EH chondrites (Consolmagno et al. 2008) are outlined next to the legend. Example screen captures of shape models used for volume determination are shown below the graph. The bars underneath the sample labels correspond to 1 cm.

This technique allowed us to identify samples with anomalous densities, leading to improved measurement precision for the current data set. A side benefit is the routine documentation of original sample shapes before some of the meteorites are cut and distributed to private collectors or used in scientific studies. The density results were compared to representative data for H, L, and EH chondrites (Consolmagno et al. 2008) in Fig. 1. We found no systematic influence on the density results from the variable numbers of scans needed to acquire the complete volumes when analyzing sample masses of 30–360 g. The measured densities of fragments from the Tamdakht and Huaxi meteorites are consistent with previous density measurements on smaller fragments, performed by El Abassi et al. (2013) and Li et al. (2017), respectively, which suggest that the bulk density data presented here are accurate.

Pre-Atmospheric Sizes

The pre-atmospheric sizes of meteorites and sampled shielding depths are important parameters for interpreting the irradiation history from samples studied using cosmogenic nuclides. In this context, the two isotopes ^{26}Al and ^{60}Co are especially useful due to their well-constrained production rates. While ^{26}Al generally is detectable in meteorites, ^{60}Co , which is mainly produced by thermal neutrons, is easier to detect in samples which have been delivered from larger meteoroids ($r > 20$ cm) and sufficient shielding depths (e.g., Spergel et al. 1986). In commonly sized chondrite meteoroids ($r < 50$ cm), the production rate of ^{26}Al increases with shielding depth down to ~ 40 cm (Leya and Masarik 2009). Pre-atmospheric size estimates for the Lost City, Tamdakht, Huaxi, Boumdeid, and Kamargaon meteorites are available based on data given in previous studies. The pre-atmospheric radius of the Lost City meteorite was less than 25 cm based on low activities of ^{60}Co and other radionuclides that were determined through gamma-ray spectrometry measurements on several different fragments shortly after the fall (e.g., Evans et al. 1982). Tamdakht was delivered by a medium-sized meteoroid with a radius of ~ 47 cm according to radionuclide measurements and simulations by Alexeev et al. (2012). The radionuclide data from Tamdakht are consistent with our measurements and shielding-dependent variations of ^{60}Co and ^{26}Al production rates. Huaxi and Kamargaon came from considerably smaller meteoroids according to calculations based on the noble gas contents in Huaxi giving a radius of ~ 11 cm (Li et al. 2017) and radionuclide activities in Kamargaon giving $r \leq 10$ cm according to Ray et al. (2017). While our data are consistent with the estimates for Huaxi, the considerably higher activity of ^{26}Al in Kamargaon measured by us contradicts the previous size estimate. A small pre-atmospheric size ($r < 20$ cm) was inferred from radionuclide data for Boumdeid (2011) in a previous study (Buhl et al. 2014) which is consistent with the results obtained by us. From the remaining meteorite fall samples, the determined maximum activity of ^{60}Co in Oudiyat Sbaa, < 0.4 dpm per kg, and the very low activity detected in Degtevo (0.3 ± 0.1 dpm per kg), along with the respective activities of ^{26}Al , suggest that these two meteorites came from pre-atmospheric bodies with radii smaller than 20 cm. The same constraints apply to the find SaU 606 which gave a low activity of ^{26}Al and, in spite of a strong signal for ^{22}Na , shows no detectable ^{60}Co . The indicated mean shielding depth of just a few cm is consistent with the fact that later systematic search for additional samples near the 2017 find site did not yield any paired stones. Although

$\sim 1\text{--}2$ dpm per kg of ^{60}Co in the samples from the Xining meteorite indicates a relatively low flux of thermal neutrons (Spergel et al. 1986), reports of the recovered mass exceeding 100 kg and bulk densities of ~ 3.4 g cm $^{-3}$ exclude a pre-atmospheric radius smaller than 20 cm when assuming a spherical object. Considering these parameters, the Xining samples likely came from relatively low shielding depths in an object with $r > 20$ cm. An expected steep increase of ^{60}Co production with increasing meteoroid radius, even at shielding depths below 5 cm (Eberhardt et al. 1961; Spergel et al. 1986), and the activities of ^{26}Al exceeding 55 dpm per kg in the Xining samples along with considerations of the corresponding production rates (Leya and Masarik 2009) limit the pre-atmospheric size of the Xining meteorite to an object with a radius of $\sim 25\text{--}65$ cm. The measured activity of ^{26}Al in the Mürtschenstock main mass, ~ 50 dpm per kg indicates that this meteorite either came from a small meteoroid ($r = 10\text{--}20$ cm) or a considerably larger object with $r < 100$ cm. Given the clear predominance of impactors in the smaller size range (e.g., Brown et al. 2002), and that no other pieces of Mürtschenstock have so far been found, a small ($r = 10\text{--}20$ cm) pre-atmospheric size of this meteorite can be considered more likely.

Repeated Measurements of Huaxi and Xining Subsamples

We measured two individual samples from each of the Huaxi and Xining meteorites, respectively. The inferred small pre-atmospheric size of the Huaxi meteorite (Li et al. 2017) combined with the low total amount of recovered mass (1.6 kg) should not allow for any significant shielding-dependent variations of cosmogenic radionuclide activities between two different fragments. This is clearly reflected in our results on Huaxi as the activities of all detected isotopes in both studied samples agree within uncertainties. The high uncertainties of ^{54}Mn are mainly due to low count rates after 6 years of decay after the fall. Measured activities of ^{40}K are identical for both samples and the corresponding K concentrations are in good agreement with a previous ICP-MS measurement performed on a smaller subsample from the same meteorite (Li et al. 2017). In the Xining meteorite ($\sim 25\text{--}65$ cm pre-atmospheric radius), activities of ^{26}Al and more short-lived ^{60}Co and ^{22}Na are not distinguishable between the two different samples which suggest that both fragments stem from similar shielding depths. The two different measurements give a large range of uncertainty for the activity of ^{54}Mn but do not provide evidence for significant variations in the production rates of this isotope. The different activities of ^{40}K in the two Xining samples do, however, suggest that K is unevenly

distributed within the sampled mass range, 30–40 g, in this meteorite. Together, the results from repeated measurements on subsamples of the Huaxi and Xining meteorites show good measurement reproducibility.

Bulk Potassium in Ordinary and Enstatite Chondrites

Potassium concentrations have often been used as a quality control when assessing the precision of gamma spectrometry measurements on ordinary chondrites (e.g., Weber et al. 2017). Occasionally, studies have adopted published average bulk values of K in corresponding chondrite groups for internal standardization. Concentrations of K in chondrite groups do, however, vary within and between bulk samples, which are evident from several series of measurements using various analytical methods, often performed on sample masses of less than 1 g. The largest available data set consists of more than 1100 wet chemistry analyses of Antarctic meteorite finds, out of which ~1000 are non-carbonaceous chondrites (Kimura et al. 2018). This data set is further distinguished by relatively large sample masses, generally more than 2 g, and that sampled meteorites represent the actual distribution of finds, unbiased by selection based on petrographic type or availability. These measurements give lower average values for K concentrations in H and L chondrites, ~690 and ~770 ppm, respectively, than previous data series (Wasson and Kallemeyn 1988; Jarosewich 1990) and the values appear normally distributed for both L and H chondrites (Fig. 2). Our measured concentrations in seven bulk samples from six different H-chondrites are in perfect agreement with the concentrations measured by Kimura et al. (2018), yielding the higher precision expected from the larger sample sizes, ~100 to 2100 g (Fig. 2). The concentration of K in Oudiyat Sbaa is lower than in average H chondrites and lower than values for EH chondrites by Wasson and Kallemeyn (1988) but fully consistent with the range of concentrations in the handful of EH chondrites samples measured by Kimura et al. (2018). Concentrations of K in Antarctic L chondrites subsamples have a larger variance, which likely can be explained by the commonly observed occurrences of shock melts and brecciation in this chondrite group. These features could potentially also explain the inhomogeneous distribution of K in Xining. In spite of additional samples needed in order to obtain a representative L chondrite mean, our results on L chondrites are consistent with the compositional distribution of Antarctic meteorite finds which further suggest that average concentrations of K delivered to the Earth through L and H chondrites are represented in the GeMSE measurements with a good accuracy and precision.

Fig. 2. Bulk K concentrations in samples measured with GeMSE compared to concentrations by Kimura et al. (2018), black bins, 100 ppm bin width. Dpm per kg for ^{40}K (Table 3) corresponds to $1.86\times$ concentration (ppm). Higher concentrations by Jarosewich (1990) overlain for comparison, gray bins, 100 ppm bin width. Oudiyat Sbaa (EH) with lower bulk concentration of K than average H chondrites is shown in the same diagram. Potassium concentrations in the two different Xining fragments are labeled. Two clear outliers in the Antarctic H chondrite data (right) set are from brecciated samples (Kimura et al. 2018).

Solar Modulation and Radionuclide Production Rates

Detected activities of cosmogenic radionuclides with very short half-lives ($t_{1/2} = 16\text{--}53$ d) in the samples from Oudiyat Sbaa (^7Be , ^{31}Cr , ^{48}V), Degtevo (^7Be), and Kamargaon (^7Be) in addition to ^{58}Co , ^{56}Co , ^{46}Sc , ^{57}Co ($t_{1/2} = 70\text{--}270$ d) are consistent with previous measurements on meteorites during similar stages of the solar cycle (Evans et al. 1982) and these three meteorite's reported dates of fall. Regular fluctuations of solar activity cause periodic variations in the cosmic ray flux, that is, a solar modulation. During periods of high solar activity, the particle flux decreases in a lagging anticorrelation with the number of observed sunspots. Due to this phenomenon, the net activities of ^{46}Sc and ^{54}Mn in meteorites vary within a factor of ~2.3 whereas ^{22}Na activities vary with a factor of ~1.5 during the course of one ~11 year solar cycle (Evans et al. 1982). Long-lived ^{26}Al is not affected by these short-term variations in the GCR flux. This is clearly reflected in the $^{22}\text{Na}/^{26}\text{Al}$ activity ratios obtained from Tamdakht, Xining, Boumdeid, Degtevo, and Oudiyat Sbaa which all show the expected decrease of ^{22}Na

Fig. 3. Activity ratio, $^{22}\text{Na}/^{26}\text{Al}$, at the time of fall measured in recent meteorite fall samples, decreasing with increasing solar activity which is indicated below by the integrated sunspot count (smoothed monthly; Source: WDC-SILSO, Royal Observatory of Belgium, Brussels) during the final stage of solar cycle 24. Dashed lines indicate the average $^{22}\text{Na}/^{26}\text{Al}$ activity ratio over a solar cycle in L and H chondrites, 1.35 and 1.5, respectively (Bhandari et al. 2002).

production rates during the increased solar activity of the late stage Solar Cycle 24 (Fig. 3). The comparatively low $^{22}\text{Na}/^{26}\text{Al}$ activity ratio of Oudiyat Sbaa can likely be explained by lower concentration of the relevant target element Mg (i.e., lower Mg/Si) in EH chondrites as compared to ordinary chondrites. The $^{22}\text{Na}/^{26}\text{Al}$ activity ratios in Huaxi and Kamargaon fall outside of the expected variation and are instead, respectively, higher and lower than activity ratios normally recorded in chondrites. These anomalies are further discussed in the following section.

Irradiation Histories of Kamargaon and Huaxi

Activities of ^{22}Na and ^{26}Al in the Kamargaon meteorite (Ray et al. 2017) are lower than our results; the large discrepancy ($\sim 50\%$) of ^{26}Al is difficult to explain. The pre-atmospheric size estimate of Ray et al. (2017), $r < 10$ cm, is likely incompatible with the reported mass found (~ 25 cm $\times$ 23 cm $\times$ 20 cm in size). A similar spherical mass with average L-chondrite density corresponds to a radius of 9.5 cm and thus would have seen a low degree of atmospheric ablation. In addition, the previously reported values for ^{26}Al and ^{22}Na of ~ 40 dpm per kg and ~ 50 dpm per kg, respectively, are only produced at the very surface of a

small L chondrite meteoroid (Bhandari et al. 1993; Leya and Masarik 2009). Similarly, the pre-atmospheric size which is indicated by our measured activity of ^{26}Al ($r > 25$ cm) is inconsistent with the failure of both studies to detect ^{60}Co . The heterogeneous distribution of ^{26}Al in the two different samples of Kamargaon is too large to be an analytical artifact. This is further indicated by the large difference in $^{22}\text{Na}/^{26}\text{Al}$. Combined, the diverging observations can only be explained by a complex exposure history. Unusually low activities of ^{26}Al in Huaxi are expected from the previously determined low average cosmic ray exposure age (1.6 ± 0.1 Ma) of this meteorite (Li et al. 2017). Assuming a single-stage exposure history and symmetrical ablation of Huaxi, we calculated the corresponding average net activity of ^{26}Al after this time. We further assumed placement in the center of an H chondrite body and a pre-atmospheric size similar to that given by Li et al. (2017). With a radius of 10 cm and 4–10 cm shielding, the production rate by Leya and Masarik (2009) yield a net activity of 34.3 ± 1.3 dpm per kg after 1.6 ± 0.1 Ma exposure. Our measurements ($^{26}\text{Al} = 34\text{--}35$ dpm per kg in the combined samples) are thus in excellent agreement with the irradiation history proposed by Li et al. (2017) and fully consistent with a recovered mass of 1.6 kg.

^{44}Ti in Chondrites

Titanium-44 can potentially record time-dependent variations in solar activity, but the uncertainties connected to our measurements are apparently too large to directly resolve any existing variations since the Lost City meteorite fall 1970. Normalized to average main target element (Fe+Ni) concentrations in the respective chondrite groups, our measurements generally give modes corresponding to ^{44}Ti activities of ~ 2 to 3 dpm per kg(Fe+Ni). The calculated mode for the Tamdakht meteorite corresponds to a higher activity of ~ 4.5 dpm per kg(Fe+Ni), but the difference is only minor within uncertainties. These values are considerably lower than the average shielding corrected activity, ~ 4.2 dpm per kg(Fe+Ni), in the youngest meteorite falls given in an earlier study (Taricco et al. 2006) that suggested a continuous decrease of ^{44}Ti production rates during the last two and a half centuries. The reason for this difference is not clear, but we cannot exclude systematic effects derived from different methods of activity calculations. The preceding study used ^{40}K as internal standard for detection efficiencies on all samples except the Torino meteorite from which a replicate mold was used as a standard geometry. Their stated concentrations of K are generally higher than those measured in GeMSE, so the derived activities of ^{44}Ti

Table 4. Activity ratios in meteorite falls (at the time of fall) calculated for data obtained in this study and samples measured with the spectrometer GEM 90 by Taricco et al. (2006). The average concentrations of Fe + Ni in chondrites (Wasson and Kallemeyn 1988), which were used to normalize the GeMSE ^{44}Ti data to the concentrations of target elements are included together with ^{26}Al scaling factors used for all data.

GeMSE		Taricco et al. (2006) recalculated data	
Falls 1970–2016	$^{44}\text{Ti}(\text{Fe}+\text{Ni})/^{26}\text{Al}$	Falls 1976–2001	$^{44}\text{Ti}(\text{Fe}+\text{Ni})/^{26}\text{Al}^b$
Oudiyat Sbaa	$0.060^{+0.017}_{-0.021}$	Dhajala	0.069 ± 0.012
Degtevo	$0.061^{+0.008}_{-0.012}$	Torino	0.085 ± 0.014
Kamargaon	$0.044^{+0.012}_{-0.019}$	Mbale A	0.060 ± 0.006
Xining A	<0.130	Mbale T	0.041 ± 0.005
Xining B	<0.120	Fermo	0.060 ± 0.005
Boumdéid	<0.080	Dergaon	0.036 ± 0.006
Huaxi A ^a	$0.045^{+0.018}_{-0.013}$	Mean	0.058 ± 0.018
Huaxi B ^a	$0.045^{+0.018}_{-0.019}$		
Tamdakht	$0.080^{+0.021}_{-0.022}$	Calculation parameters	
Lost City	$0.043^{+0.022}_{-0.019}$	Avg. Fe+Ni in H	29.1 wt%
Finds		Avg. Fe+Ni in L	22.7 wt%
SaU 606	$0.063^{+0.023}_{-0.030}$	Avg. Fe+Ni in EH	30.75 wt%
Mürtschenstock	<0.080	$^{26}\text{Al}(\text{bulk}) \text{ L/H}$	1.08
Mean (detected)	0.055 ± 0.013	$^{26}\text{Al}(\text{bulk}) \text{ LL/H}$	1.09

These and the concentrations of Fe+Ni which were used for target normalization of activities measured by Taricco et al. (2006) were taken from the original publication.

^aAdjusted to the average saturation activity of ^{26}Al ($r = 10, 4\text{--}10$ cm shielding depth).

^bRecalculated $^{44}\text{Ti}/^{26}\text{Al}$ using the efficiency ratio of Torino with uncertainties corresponding to the count rates.

are likely to diverge. For easier comparison of the two data sets, we assess the $^{44}\text{Ti}(\text{Fe}+\text{Ni})/^{26}\text{Al}(\text{bulk})$ activity ratios from the different measurement series. Most of our measurements were performed on H chondrites so to evaluate corresponding production rates in other meteorite types we correct for the difference between chondrite groups expected from the concentration of target elements and production rates of ^{26}Al . For this, we use the calculated average production rates of ^{26}Al in L and LL compared to H (factor of 1.08 and 1.09, respectively) in accordance with Leya and Masarik (2009). For Oudiyat Sbaa, we assume a production rate corresponding to H chondrite compositions. When treating the data by Taricco et al. (2006), we used their given measurement data (count rate, activity, and ^{44}Ti decay time) from the sample of the Torino meteorite to calculate the detection efficiency ratio $\epsilon^{44}\text{Ti}/\epsilon^{26}\text{Al}$. Under the assumption that this ratio should be similar for all samples which were measured with the same spectrometer, we then recalculate the corresponding $^{44}\text{Ti}(\text{Fe}+\text{Ni})/^{26}\text{Al}$, using the Torino efficiency ratio and the measured values for Fe and Ni concentrations for meteorites which fell after 1970 (Table 4). This approach might introduce unaccounted uncertainties compared to the GeMSE data since the previous study measured ^{44}Ti in coincidence counting with the positron annihilation line at 511 keV. After recalculation, the $^{44}\text{Ti}(\text{Fe}+\text{Ni})/^{26}\text{Al}$ data obtained in GeMSE are statistically indistinguishable from earlier measurements

on meteorite falls, 1976–2001 (Fig. 4). The $^{44}\text{Ti}(\text{Fe}+\text{Ni})/^{26}\text{Al}$ activity ratios from both ours and the previous measurements show no apparent correlation with bulk activities of ^{26}Al which were scaled to H chondrite production rates (Fig. 4a). This is surprising since it might suggest relatively constant $^{44}\text{Ti}/^{26}\text{Al}$ production ratios in small chondrites, which would contrast with the fairly constant activities of ^{44}Ti suggested by the production model of Michel and Neumann (1998) combined with the well-known increase of ^{26}Al production with increased shielding depth in small meteoroids. Due to its complicated exposure history, the $^{44}\text{Ti}/^{26}\text{Al}$ production ratio suggested by the Kamargaon meteorite data might be underestimated and, as the activity of ^{26}Al in this meteorite does not appear to reflect its late irradiation history, the indicated pre-atmospheric size may not be correct. Whether activities of ^{44}Ti below the production rates anticipated by Michel and Neumann (1998) in recent falls are caused by century-scale variations in solar activity is beyond the scope of the current study, but our results combined with the recalculated earlier data do not provide any evidence for decreasing production rates during the last five decades. Anchored to target element concentrations and ^{26}Al production rates in H chondrites, the GeMSE measurements give a mean $^{44}\text{Ti}(\text{Fe}+\text{Ni})/^{26}\text{Al}$ of 5.5×10^{-2} with an uncertainty of 24% 1RSD. Deviations from the mean are, however, generally accounted for by the large

Fig. 4. a) Activity ratios $^{44}\text{Ti}(\text{Fe}+\text{Ni})/^{26}\text{Al}$ from GeMSE (this study) and recalculated activity ratios in previously analyzed 1976–2001 meteorite falls (Taricco et al. 2006) against the bulk activity of ^{26}Al (fixed mean/mode, uncertainties are not depicted), scaled to H chondrite production rates. b) Same analytical data as (a) showing the time when the meteorites fell. Lines indicate the average mean value of falls measured in GeMSE (black) and one standard deviation (dashed gray).

uncertainties of single measurements. From these data, the terrestrial age of a chondrite with a pre-atmospheric size range of ~10–50 cm radii can, after positive detection of ^{44}Ti , be estimated by:

$$\text{Age}_{T(44\text{Ti})} = \ln\left(\frac{A_{44\text{Ti}(\text{Fe}+\text{Ni})}}{A_{26\text{Al}} \times 0.055}\right) \times \frac{-1}{\lambda_{44\text{Ti}}} \quad (1)$$

where $A_{44\text{Ti}}$ is the activity of ^{44}Ti expressed in dpm per kg(Fe+Ni), $A_{26\text{Al}}$ is the bulk sample activity of ^{26}Al in dpm per kg scaled to an H chondrite composition, and $\lambda_{44\text{Ti}}$ is the decay constant of ^{44}Ti . The current activities of ^{44}Ti in freshly fallen bulk H chondrites appear to be consistent at ~0.5 to 1 dpm per kg and recurrently 50% lower than modeled production rates. The fact that our results were obtained through measurements of several chondrite samples with different concentrations of target elements, masses of 100–2000 g, and verified terrestrial ages of 0–47 years combined with similar results derived from previously published data strengthens this interpretation. The consistency of the results further suggests that $^{44}\text{Ti}/^{26}\text{Al}$ activity ratios in chondrites obtained with GeMSE may be regarded as valid down to the predefined Bayesian statistical limit of positive evidence for detection of ^{44}Sc . At such low

activities, ~0.5 to 1 dpm per kg, terrestrial age determinations of bulk ordinary chondrites through gamma-ray spectrometry measurements of ^{44}Ti require large sample masses and/or longer counting times; the ability to resolve a signal is less dependent on intrinsic background levels than Compton scattering from prominent isotopes in the meteorite samples. The more short-lived cosmogenic isotope ^{22}Na can be detected at >0.5 to 1 dpm per kg up to ~20 years after a meteorite fall. Due to the well-constrained average $^{22}\text{Na}/^{26}\text{Al}$ production ratios in chondrites and the large uncertainties of our ^{44}Ti measurements, the practical usefulness of the $^{44}\text{Ti}(\text{Fe}+\text{Ni})/^{26}\text{Al}$ activity ratio is generally limited to chondrite finds in which ^{22}Na can no longer be detected. In suitable cases, we approximate that this can be applied to chondrites with terrestrial ages of ~20 to 300 years.

The Terrestrial Ages of SaU 606 and Mürtchenstock

Based on the detected activities of short-lived cosmogenic radionuclides, ^{22}Na and ^{54}Mn , SaU 606 most likely fell sometime during 2012 but direct comparison with other measured fall samples can potentially put further constraints on the terrestrial age of this meteorite. The $^{22}\text{Na}/^{26}\text{Al}$ activity ratio is

generally considered to be practically invariant with shielding depth in chondrites (e.g., Bhandari et al. 2002). From this assumption, we expect the $^{22}\text{Na}/^{26}\text{Al}$ activity ratio in chondrites with similar compositions and different dates of fall to vary as a function of the average $^{22}\text{Na}/^{26}\text{Al}$ production ratio during the time between two different falls. Figure 5a depicts the maximum range for average activities in meteorites during the time between the Huaxi and Degtevo meteorites dates of fall, under the assumption that the sampled net production of ^{22}Na decreased with an increase of solar activity during the last ~ 2.5 half-lives of this radionuclide at a rate which adds up to the activity ratio in Degtevo at the time of fall. Compared to the decay curve of ^{22}Na , which corresponds to the $^{22}\text{Na}/^{26}\text{Al}$ ratio measured in SaU 606, this gives possible fall dates between early April 2012 and late May 2013. The later dates are however unlikely since they would correspond to the uppermost and lower most range of uncertainty in Huaxi and SaU 606, respectively. This suspicion is confirmed by the corresponding curves for ^{54}Mn decay in SaU 606 assuming that the activity of ^{54}Mn in SaU 606 was not lower than the lowest uncertainty range given by Evans et al. (1982) for meteorites that fell during the course of solar cycle 20 (Fig. 5b). The possible range of fall dates is thus further limited to sometime between April and December 2012, with mean values giving a best estimate of July–September 2012. In the case of Mürtschenstock, only an upper limit for the activity of ^{44}Ti could be determined. With <0.8 dpm per kg, this upper activity limit is too high to be used for a terrestrial age estimate according to activities measured in recent falls, so for a minimum terrestrial age of this sample, we instead use the determined maximum $^{22}\text{Na}/^{26}\text{Al}$ activity ratio (Table 3). Replacing the activity ratio and decay constant in (1) with maximum $^{22}\text{Na}/^{26}\text{Al}$ in Mürtschenstock and $\lambda_{^{22}\text{Na}}$, the average $^{22}\text{Na}/^{26}\text{Al}$ activity ratio, 1.35, in L chondrite falls correspond to a minimum terrestrial age of 19 years prior the find of the Mürtschenstock meteorite. The fall thus occurred in the past millennium and is not connected to the bolide observation on March 15, 2015.

Cesium-137 as a Potential Geographic Tracer

Cesium-137, produced in nuclear weapons tests in the 1950s, is now detectable in soil samples from most areas of the world. Due to the Chernobyl accident in 1986, ^{137}Cs soil activities are elevated in European soils (e.g., Szegvary et al. 2007) when compared to Arabia or northern Africa. In Oman hot desert meteorite finds, ^{137}Cs has been detected, but the determined activities were only <0.2 to 10.3 dpm per kg (median 1.25 dpm per kg) and with lower bulk activities in large masses

(Weber et al. 2017). In this context, it should be noted that contamination of ^{137}Cs likely is concentrated to the exposed surface of meteorites, so activity values obtained with gamma-ray spectrometry are not fully representative for concentrations in the analyzed bulk material. The absence of measurable ^{137}Cs in SaU 606, however, still suggests that meteorites need several years (>5 a) to accumulate even detectable levels of this isotope in the Oman desert. In contrast, the meteorite find Mürtschenstock from Switzerland does, in spite of a low degree of weathering, show a strong signal of ^{137}Cs ($42.2^{+3.1}_{-1.4}$ dpm per kg) which likely is the result of Chernobyl fallout present at the find site. The substantially higher activity of ^{137}Cs in Mürtschenstock compared to meteorites from Oman suggests that there exists a potential to broadly distinguish the find regions of meteorite samples with similar terrestrial ages, based on their acquired activities of ^{137}Cs .

CONCLUSIONS

Radionuclide activities were measured in samples from eight chondrite falls (1970–2016) and two finds. Bulk densities of meteorite samples which were accurately determined from optical shape models proved useful for the correct implementation of sample geometries in Monte Carlo-based efficiency calculations. Measured bulk concentrations of K in several kilograms of equilibrated ordinary chondrites are consistent with recently published data obtained from aliquots of more than 1000 Antarctic meteorites but often lower than average concentrations given in earlier data compilations. Most of the studied samples had cosmogenic radionuclide activities similar to previous observations from meteorites derived from small- to medium-sized pre-atmospheric bodies. The unusually low activity of ^{26}Al in the Huaxi meteorite is fully consistent with its previously determined exposure age making the irradiation history of this meteorite unusually well constrained. The erratic cosmogenic nuclide characteristics of the Kamargaon meteorite suggest a complex exposure history. Detected activities of ^{44}Ti in samples from small recent meteorite falls seem consistent with previous $^{44}\text{Ti}/^{26}\text{Al}$ count ratios obtained from meteorite falls after 1970 and further suggest that the current activities of this isotope in meteorite falls are substantially lower than modeled production rates. Results from our study give average $^{44}\text{Ti}(\text{Fe}+\text{Ni})/^{26}\text{Al}$ of 0.055 with 1RSD of 24% in H chondrites derived from small- to medium-sized meteoroids and allow for absolute or minimum terrestrial ages to be determined although the precision is limited by high measurement uncertainties. For bulk chondrite samples, the positive detection of ^{44}Ti generally requires large sample masses

Fig. 5. a) Net $^{22}\text{Na}/^{26}\text{Al}$ activity curves (whole drawn lines) over time for average production ratio of $^{22}\text{Na}/^{26}\text{Al}$ (P). $P = 1.25$ corresponds to the time between the fall of Huaxi and Degtevo. Saturated production rates of ^{26}Al , corresponding to the exposure age given by Li et al. (2017), were assumed for Huaxi. Line from the mean ratio in Tamdakht, corresponding to $P = 1.20$, is shown for comparison. The decay curve (dashed lines) for $^{22}\text{Na}/^{26}\text{Al}$ in SaU 606 intersects with the net activity, constrained by the preceding and following meteorite falls, at a time between April 2012 and May 2013. b) Constraints from measured ^{54}Mn dpm per kg(Fe+Ni). Prior and later falls are shown for reference. For the absolute minimum of ^{54}Mn activity, normalized to average concentrations of Fe+Ni, given by Evans et al. 1982 (140 dpm per kg(Fe+Ni)) the fall of SaU 606 could not have occurred after the shift 2012–2013. This leaves a possible time range of April 2012 to late December 2012 for the SaU 606 meteorite. The combined mean values give a most likely time of fall between July and September 2012.

and/or several weeks screening time even at very low intrinsic background levels. In such ideal cases, we estimate that ^{44}Ti can be useful in bridging the gap between terrestrial ages from short-lived ^{22}Na (up to ~20 a) and long-lived ^{14}C (>500–1000 a) in a time range of 20–300 years after fall. We could not detect ^{44}Ti in Mürtschenstock and the upper activity limit was too high to determine a minimum terrestrial age. The maximum $^{22}\text{Na}/^{26}\text{Al}$ activity ratio instead gives a minimum terrestrial age for the Mürtschenstock meteorite corresponding to 19 years prior to its find which rules out any connection between this meteorite and the bolide observation on March 15, 2015. Remnant activities of ^{22}Na and ^{54}Mn measured in SaU 606 compared to other H chondrites with known dates of fall suggest that this meteorite was derived from a small meteoroid that fell in July–September 2012. The substantially higher counts of ^{137}Cs in the analyzed Mürtschenstock main mass compared to hot desert finds are likely due to contamination acquired in an area which was affected by fallout from the Chernobyl accident.

Acknowledgments—This study was supported by the Swiss National Science Foundation (SNF) grant CR3312_152941. Recovery of SaU 606 was made in the frame of a project supported by SNF grant 200020_166251. We thank the Public Authority for Mining (Muscat, Sultanate of Oman) for supporting the Omani-Swiss meteorite search project. We are grateful to Aditi Bezbaruah, Abdelaziz Habibi, Tony Irving, Marc Jost, Shijie Li, Dipankar Pathak, Andreas Stucki, and Tim McCoy for kindly making meteorite samples available for our study. We thank Diego Ramirez Garcia, Jean-Michel Vuilleumier, and AEC/LHEP Bern for supporting the operation of GemSE.

Editorial Handling—Dr. Marc Caffee

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