EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Production of ^{10}Be and ^{26}Al radionuclides via neutron reactions in SiO_2 rocks: implications for burial dating related to the Cradle of Humankind–UNESCO World Heritage Site

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Abstract: The production of ^{10}Be and ^{26}Al through neutron reactions in SiO_2 (quartz-bearing) rocks plays a crucial role in cosmogenic nuclide burial dating, which determines the time since a rock or sediment was shielded from cosmic radiation. In this study, we focus on the case of the Sterkfontein Cave, where understanding the production rates of these radionuclides is essential for refining burial dating techniques. We propose to investigate the neutron-induced production of ^{10}Be and ^{26}Al by exposing quartz-bearing rock samples to the neutron flux at the EAR1, EAR2, and NEAR stations.

Requested protons: 8.0×10^{18} protons on target
Experimental Area: EAR1 (parasitic irradiation), EAR2 (parasitic irradiation) and NEAR Station (dedicated irradiation)

1 Introduction

The Cradle of Humankind UNESCO World Heritage Site (COHWHS) in South Africa is a karstic dolomite region known for its extensive cave systems and remarkable fossil discoveries. Among them, the Little Foot skeleton, dated to approximately 3.67 million years, and Homo Naledi, discovered in 2013 and dated to around 300 thousand years, provide critical insights into early human evolution. These fossils are embedded within rock layers that have preserved biological evidence for millions of years.

Geo-dating relies on measuring the concentration of long-lived isotopes such as ^{26}Al ^{10}Be in SiO_2-rich rocks [1], as their ratio provides an estimate of the sample's age (typical reference methods apply). These isotopes are primarily produced either by cosmic ray fast muons slowed down in crossing several meters of rock material and then being captured, or by cosmic-ray fast neutrons interacting by spallation with surface minerals. However, in the latter case, current uncertainties in the nuclear cross-sections used to interpret isotope concentrations result in significant dating uncertainties.

To improve accuracy of geo-dating, the best approach involves:

- Direct measurement of ^{26}Al and ^{10}Be production rates using fast neutrons above 15 MeV—the energy threshold for these nuclear reactions in silicon and oxygen—through experiments.
- In situ measurement of the fast neutron flux above this energy threshold in COHWHS cave systems to refine dating models.

These studies will enhance the precision of geo-dating methodologies, helping to establish a more reliable timeline for fossil-bearing rock formations in the region. In particular, direct measurements of the ^{26}Al and ^{10}Be production rates using fast neutrons can be performed at CERN's n_TOF facility.

2 Burial dating

Cosmogenic nuclide burial dating is becoming increasingly important in archaeology and paleoanthropology. As a dating method applicable over the past 5 million years, burial dating fills a crucial niche in these fields. It holds particular promise for dating Earlier Stone Age (ESA) sites and for determining the timing of hominin presence outside East Africa.

Cosmogenic nuclide burial dating is based on the radioactive decay of extremely rare nuclides produced in rocks by cosmic rays. These nuclides accumulate in rocks exposed near the Earth's surface. When the rocks are subsequently buried—such as in a cave or an alluvial deposit—cosmic rays can no longer reach them, and the cosmogenic nuclide inventory begins to decay over time [1, 2].

Although a variety of cosmogenic nuclides are produced, only a few can be used in geochronology. To be useful, cosmogenic nuclides must be distinguishable from any naturally occurring isotopes in the rock and must be produced in measurable quantities. Table 1 lists commonly used cosmogenic nuclides along with their radioactive half-lives.

The in situ-produced cosmogenic nuclides that have been most commonly used in archaeology and paleoanthropology are primarily ^{10}Be and ^{26}Al, both being produced in the mineral quartz. Quartz is the ideal mineral for cosmogenic nuclide dating for several reasons. First, it is widely available and highly resistant to weathering, making it easy to find in many environments. Second, quartz has a simple, stable chemistry, which allows for precise detection of cosmogenic

Nuclide	Primary Targets	Half-life	Primary production mechanisms
^{10}Be	O, C	1.39 Ma	Spallation, μ^- capture
^{14}C	O	5730 yr	Spallation, μ^- capture
^{26}Al	Si	0.71 Ma	Spallation, μ^- capture
^{36}Cl	Ca, K, ^{35}Cl	0.3 Ma	Spallation, μ^- capture, n capture
^{3}He	All	Stable	Spallation
^{21}Ne	Si	Stable	Spallation, μ^- capture

Table 1: Nuclides produced by cosmic rays, used for geochronology.

nuclides like ^{10}Be and ^{26}Al, even in low concentrations. Third, quartz can be easily separated from other minerals and cleaned of contaminants, ensuring accurate results. Finally, it's often abundant at archaeological sites, either as part of the sediment or as tools and artifacts, making it especially useful for archaeological research.

Quartz-bearing rocks, soils, and sediments at the surface accumulate ^{10}Be and ^{26}Al over time due to exposure to cosmic rays, in particular neutrons. While the exact amount in any grain is unpredictable due to variations in exposure history, both nuclides share the same history, meaning their ratio follows a defined equation. When quartz is suddenly buried—whether in a cave, river terrace, colluvial wedge, or under cultural deposits—its inherited nuclide concentrations begin to decay, following this equation:

$$N_i(t) = N_{i,inh}\, e^{-t/\tau_i} + N_{i,pb} \tag{1}$$

where the subscript *inh* denotes inheritance (i.e., the amount of cosmogenic nuclides at the time of burial), and the subscript *pb* denotes postburial production, while index i refers to the isotope of interest. For the site of interest, the rocks containing fossils to be dated have an ^{10}Be and ^{26}Al concentration, measured using AMS techniques, ranging from $5 \cdot 10^5$ to $1 \cdot 10^6$ atoms/g and from $8 \cdot 10^5$ to $5 \cdot 10^6$ atoms/g, respectively.

The key to burial dating is solving equation 1 simultaneously for ^{10}Be and ^{26}Al.

$$(N_{26} - N_{26,pb})/(N_{10} - N_{10,pb}) = R_{inh}\, e^{-t/\tau_{bur}} \tag{2}$$

where $\tau_{bur} = 1/(1/\tau_{26} - 1/\tau_{10})$ is a decay constant and $R_{inh} = (N_{26,inh}/N_{10,inh})$[2].

Equation 2 is central to the burial dating method, as all related approaches rely on its simplifications or modifications. There are three main methods that simplify the equation and are used for geodating. The first is the so-called Simple Burial Dating method, which assumes post-burial nuclide production to be negligible. The second is the so-called Min/Max Burial Dating method, where the minimum is obtained with the same assumptions as the Simple Burial Dating, while the maximum is estimated considering the post-burial production using muon production profiles. The third approach is the so-called Isochron Burial Dating method, which relies on multiple samples to explicitly determine post-burial production. To date the fossil-bearing rocks at the site of interest for our project, the third method cannot be applied, so it is necessary to focus on the first two.

In simple burial dating, the samples are buried deeply enough that post-burial production by muons can be ignored. Therefore, Equation 2 simplifies to

$$N_{26}/N_{10} = R_{inh}\, e^{-t/\tau_{bur}} \tag{3}$$

Considering $\tau_{10} \approx \tau_{26}$, R_{inh} can be calculated and the Equation 3 becomes

$$N_{26}/N_{10} = [(P_{26}/P_{10})/(1 + N_{10}^{*}\, e^{t/\tau_{10}})]\, e^{-t/\tau_{bur}} \tag{4}$$

where P_{10} and P_{26} are the production rates at the surface of ^{10}Be and ^{26}Al, $N_i^* = N_i/(P_i\tau_i)$ is the concentration normalized to the secular equilibrium, $\tau_{bur} = 1/(1/\tau_{26} - 1/\tau_{10})$ and τ_{10} and τ_{26} are the decay constant of ^{10}Be and ^{26}Al.
The Min/Max method is used when the samples to be dated have not been buried deep enough to ignore post-burial production. In this case, the minimum burial age is calculated using the Simple Burial method, while the maximum is determined by considering the highest post-burial production rates that the sample could have experienced.
For both methods, a key parameter to define is the production rate: at the surface for the Simple Method and at the burial depth of the sample for the post-burial production rate, which is then used to calculate the maximum burial age. The production rate in the literature is calculated by combining the different processes that cause the production of the isotopes of interest, namely:

- spallation due to the interactions between neutrons in secondary cosmic rays and rocks (the typical neutron spectrum at the surface is shown in Figure 1);
- interactions between muons from cosmic rays and rock (negative muon capture and fast muon reaction).

Production rates by all three methods (neutron spallation, negative muon capture, and fast muon reactions) can be approximated by the following equation, with the variables $A_{i,j}$ and L_j representing production rate factors of reaction j in nuclide i, and penetration length factors respectively

$$P_i = \sum_j A_{i,j}\, e^{-z/L_j} \tag{5}$$

Considering the parametrization by Braucher *et al.* [3], Figure 2 shows the production rate of the ^{10}Be (left) and ^{26}Al (right) and the contribution of each process to it. At the surface, the main contribution is given by spallation due to neutrons, therefore to calculate the simple burial age it's crucial the knowledge of how neutrons interact with rocks.
Considering that the rocks at the site of interest are predominantly quartz, the possible reactions involving neutrons that produce ^{10}Be and ^{26}Al are listed in Table 2. The direct measurement of the production rate can improve dating by replacing the previously obtained value, derived through approximations, with experimental measurements.

3 Experimental campaign at n_TOF

The cross sections for the neutron-induced reactions in Tab. 2 as a function of kinetic energy would be required for an accurate estimate of the production rate of ^{10}Be and ^{26}Al. However, beyond the experimental complexity in performing such measurements, one can argue that in first approximation the neutron-induced spectrum-integrated production rate could be deduced by irradiating SiO_2 samples over the n_TOF neutron beams. In fact, in the energy region of interest, i.e. $E_n > 10$ MeV, the neutron spectra available at EAR1, EAR2, and NEAR are similar to the neutron spectrum form cosmic rays showed in Fig. 1. The difference is related to the position of the intra-nuclear cascade peak: approximately at $E_n = 200$ MeV for cosmic-ray fast neutrons [4], and $E_n = 80 - 150$ MeV at EAR1, EAR2, and NEAR. In principle, the comparison of the obtained production rates makes the extrapolation of the production rate in the energy range of interest possible. Moreover it can be used for the estimate of its uncertainty.
We propose to irradiate 2-gram SiO_2 samples in the form of quartz cylinder of 1-cm radius, thus containing 0.0064 SiO_2 molecules per barn. Samples of pure SiO_2-powder from commercial

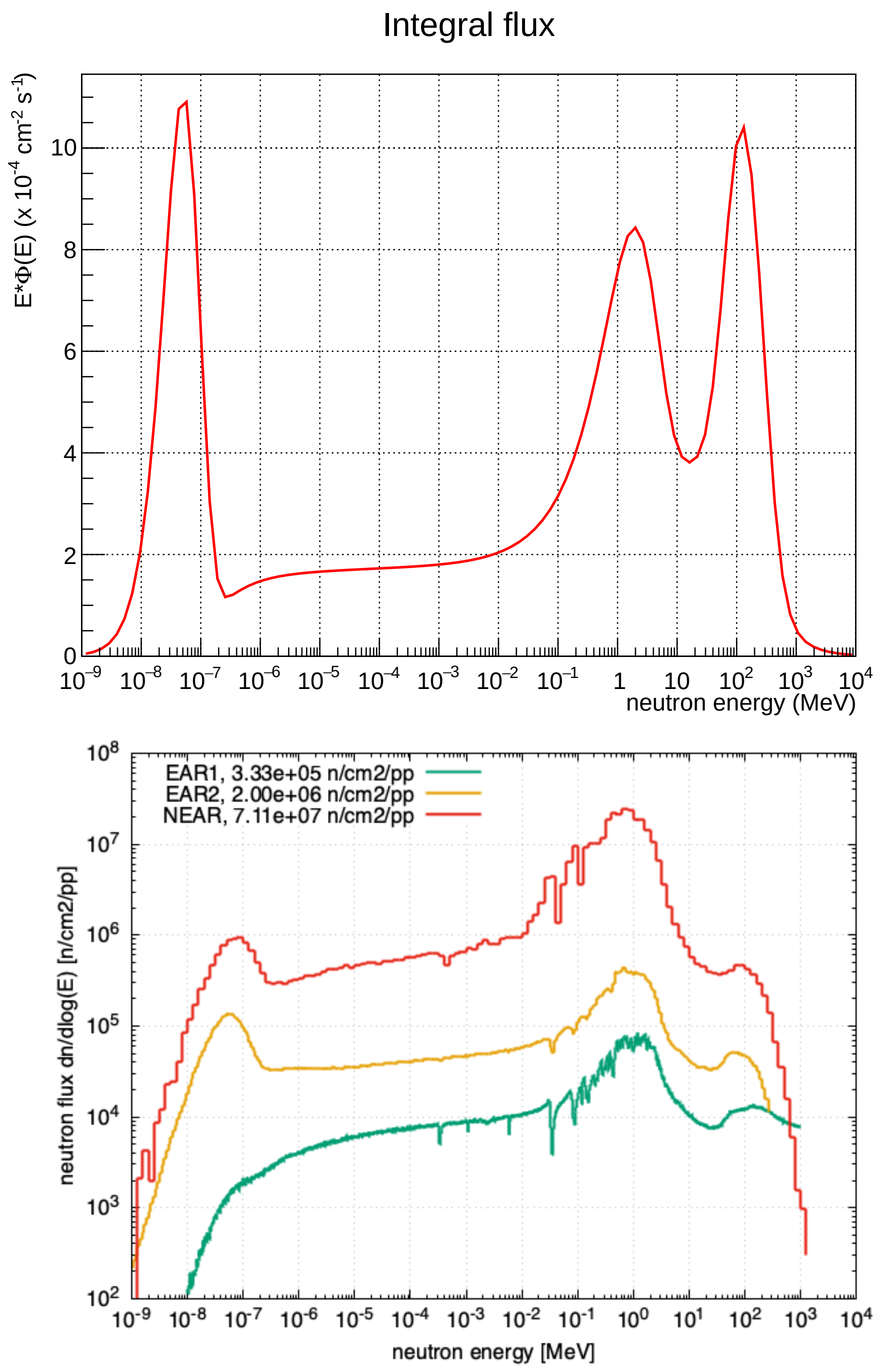

Figure 1: (upper panel) the energy distribution of neutrons from the spallation of cosmic rays in the atmosphere shows several distinct features, including peaks associated with different physical processes: thermal-energy neutrons at a few meV, an evaporation peak at a few MeV, and an intra-nuclear cascade peak at a few hundred MeV. (lower panel) energy distribution of the neutron beam at the three experimental areas of the n_TOF facility. The values shown are for each standard proton pulse (pp) of 7×10^{12} protons.

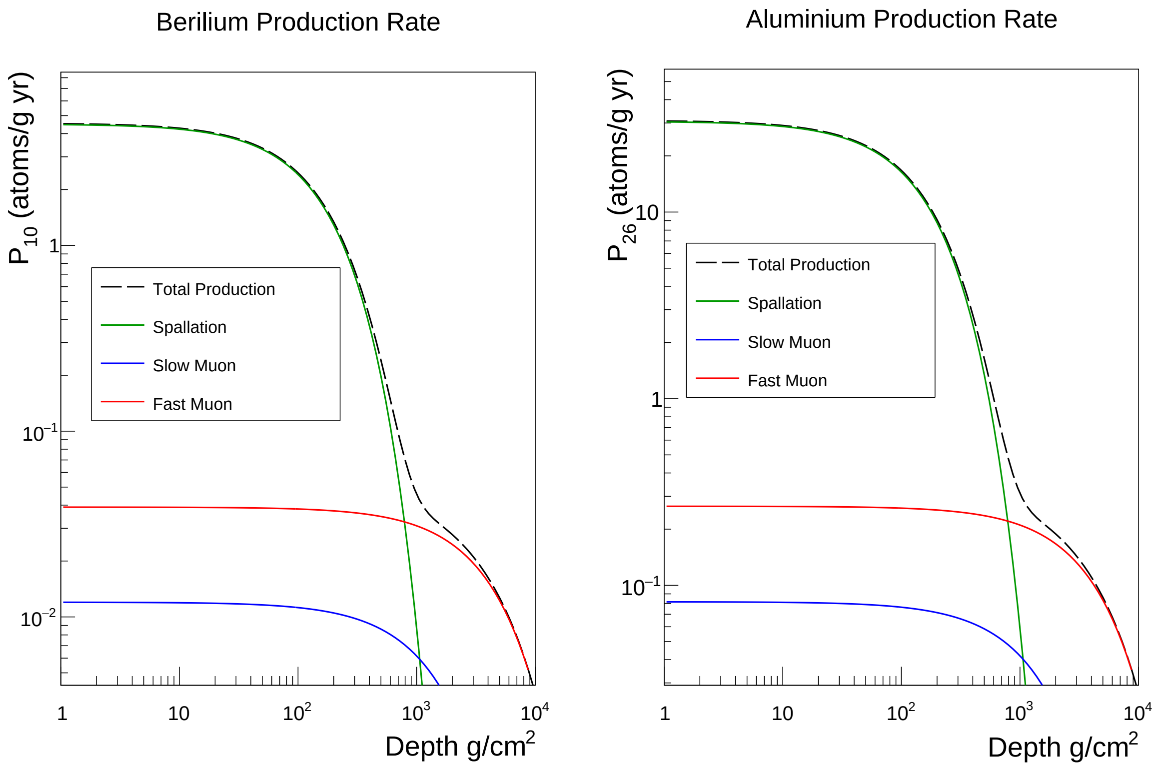

Figure 2: Production rate for ^{10}Be (left) and ^{26}Al as function of the depth in metres of water equivalent.

n + ^{28}Si			
Reaction Products	Threshold (MeV)	Reaction Products	Threshold (MeV)
$^{26}\mathrm{Al} + \mathrm{t}$	16.74	$^{10}\mathrm{Be} + {}^{3}\mathrm{He} + 4\alpha$	52.49
$^{26}\mathrm{Al} + \mathrm{n} + \mathrm{d}$	23.23	$^{10}\mathrm{Be} + \mathrm{p} + \mathrm{d} + 4\alpha$	58.18
$^{26}\mathrm{Al} + 2\mathrm{n} + \mathrm{p}$	25.53	$^{10}\mathrm{Be} + \mathrm{p} + \mathrm{t} + {}^{3}\mathrm{He} + 3\alpha$	73.02
		$^{10}\mathrm{Be} + \mathrm{n} + 2\,{}^{3}\mathrm{He} + 3\alpha$	73.81
		$^{10}\mathrm{Be} + 2\mathrm{d} + {}^{3}\mathrm{He} + 3\alpha$	77.19
		$^{10}\mathrm{Be} + \mathrm{d} + \mathrm{t} + 2\,{}^{3}\mathrm{He} + 2\alpha$	92.03
n + ^{16}O			
Reaction Products	Threshold (MeV)	Reaction Products	Threshold (MeV)
$^{10}\mathrm{Be} + {}^{3}\mathrm{He} + \alpha$	28.31	$^{10}\mathrm{Be} + \mathrm{n} + \mathrm{p} + \mathrm{d} + {}^{3}\mathrm{He}$	56.03
$^{10}\mathrm{Be} + \mathrm{p} + \mathrm{d} + \alpha$	34.14	$^{10}\mathrm{Be} + \mathrm{n} + 3\mathrm{p} + \mathrm{t}$	57.58
$^{10}\mathrm{Be} + \mathrm{n} + 2\mathrm{p} + \alpha$	36.52	$^{10}\mathrm{Be} + 2\mathrm{n} + 2\mathrm{p} + {}^{3}\mathrm{He}$	58.39
$^{10}\mathrm{Be} + \mathrm{p} + \mathrm{t} + {}^{3}\mathrm{He}$	49.37	$^{10}\mathrm{Be} + \mathrm{p} + 3\mathrm{d}$	59.50
$^{10}\mathrm{Be} + \mathrm{n} + 2^{3}\mathrm{He}$	50.19	$^{10}\mathrm{Be} + \mathrm{n} + 2\mathrm{p} + 2\mathrm{d}$	61.87
$^{10}\mathrm{Be} + 2\mathrm{d} + {}^{3}\mathrm{He}$	53.66	$^{10}\mathrm{Be} + 2\mathrm{n} + 3\mathrm{p} + \mathrm{d}$	64.23
$^{10}\mathrm{Be} + 2\mathrm{p} + \mathrm{d} + \mathrm{t}$	55.21		

Table 2: List of the possible reaction to produce ^{10}Be and ^{26}Al starting from quartz (SiO_2)

	protons ($\times 10^{18}$)			expected reactions		
Sample	EAR1	EAR2	NEAR	EAR1	EAR2	NEAR
pure SiO_2	4	2	2	$\sim 10^7$	$\sim 10^7$	$\sim 10^8$

Table 3: Proposed irradiation and order of magnitude of the expected number of nuclear reactions.

providers as well as from South Africa's rocks will be irradiated to check for the role of other contaminants. Irradiations with and without B_4C filters are needed to address the contribution of thermal and epithermal neutrons. Details are given in Tab. 3, where the number of reactions is calculated assuming a reaction cross section of 20 mb.
The number of requested protons at NEAR is calculated to reproduce the number of neutrons reaching a rock sample over 3.67 million years. For EAR1 and EAR2, the beam time is calculated to achieve a concentration of ^{10}Be and ^{26}Al that is 10 times higher than in reference samples.
While irradiations in EAR1 and EAR2 can be performed in parasitic mode (i.e., during other experimental campaigns), dedicated beam time is required at NEAR.
A sample holder will contain four samples: one of pure SiO_2, one of pure SiO_2 contained in a B_4C filter, one of SiO_2 from South Africa, and one of SiO_2 from South Africa contained in a B_4C filter. All samples will be irradiated simultaneously.
Once irradiated, the ^{10}Be and ^{26}Al radionuclide measurements will be performed using accelerator mass spectrometry.
Assuming an uncertainty of 4-5% on the AMS determination of ^{10}Be and ^{26}Al, and a 3% uncertainty on the neutron flux, we aim to estimate the production rate with an overall uncertainty of up to 6%.
Summary of requested protons: 4.0×10^{18} for EAR1 (parasitic irradiation), 2.0×10^{18} for EAR2 (parasitic irradiation), 2.0×10^{18} for NEAR (dedicated irradiation).

4 Acknowledgments

The activity described in this project is part of a broader initiative currently funded by INFN Italy, known as MUSTAR (MUons at the Rising STAR site). In addition to INFN—through its Bologna and Firenze sections and the National Laboratory of Legnaro—and other Italian partners as the University of Firenze, several other international institutions and organizations are actively involved in the initial phase of this project. This phase, a Proof of Concept, is scheduled to take place in the first half 2025 at site U.W. 105 of the Rising Star cave system.. These collaborating institutes include the Lee R. Berger Foundation (LRBF), National Geographic. Formal agreements between INFN and these institutions are in an advanced stage.
We wish to fully acknowledge the crucial contributions of these groups, both in providing rock samples from the site and in offering essential input for the planning and objectives of the proposed measurements.
We also gratefully acknowledge Dr. Tebogo Makhubela of the University of Johannesburg for his pivotal role in initiating the geodating project using cosmic rays.

References

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
SIMON at EAR1 and SIMON at EAR2	☒ To be used without any modification ☐ To be modified
SiO_2 samples	☒ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from flexible or transported equipment to the CERN site:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☐	
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	
Electrical Safety	Electrical equipment and installations	☐	
	High Voltage equipment	☐	
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	☐	
	Toxic/Irritant	☐	
	Corrosive	☐	
	Oxidizing	☐	
	Flammable/Potentially explosive atmospheres	☐	
	Dangerous for the environment	☐	
Non-ionizing radiation Safety	Laser	☐	
	UV light	☐	
	Magnetic field	☐	
Workplace	Excessive noise	☐	
	Working outside normal working hours	☐	
	Working at height	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards		☐	