Supplemental Information

Web table 1. ^{3}He and ^{4}He concentrations, ^{3}He/^{4}He ratios, and non-carbonate fraction in Gubbio limestones. He concentrations are per gram of bulk sediment and [^{3}He], [^{4}He], ^{3}He/^{4}He represent averages of leached replicates. R$_A$ is the ^{3}He/^{4}He ratio normalized to the atmospheric value of 1.39×10^{-6}. NCF = non-carbonate fraction in the sediments as measured by our mass loss data. The 1σ uncertainty is based on reproducibility of replicate analyses (*1, 2*).

Stratigraphic Height (m)	[^{3}He] 10^{-15} cc STP g^{-1}	±1σ	[^{4}He] 10^{-9} cc STP g^{-1}	±1σ	^{3}He/^{4}He R$_A$	NCF
346.45	41.9	8.1	33.1	2.9	0.9	0.05
346.50	122.8	23.3	148.1	12.9	0.6	0.17
346.55	41.3	7.8	58.3	5.1	0.5	0.07
346.60	70.2	13.6	51.0	4.4	1.0	0.06
346.65	80.1	15.6	46.9	4.1	1.2	0.06
346.70	75.9	10.3	62.5	3.8	0.8	0.06
346.75	29.9	5.6	46.7	4.1	0.5	0.06
346.80	71.4	13.9	44.1	3.8	1.2	0.06
346.85	40.0	7.5	64.4	5.6	0.4	0.07
346.90	49.6	9.5	44.4	3.9	0.8	0.06
346.95	67.9	13.0	72.7	6.3	0.7	0.08
347.00	39.0	7.3	65.1	5.7	0.5	0.07
347.10	88.2	17.2	47.5	4.1	1.3	0.06
347.15	47.8	9.1	53.2	4.6	0.6	0.07
347.25	91.1	17.9	43.6	3.8	1.5	0.05
347.40	23.0	4.2	51.8	4.5	0.3	0.05
347.45	49.9	9.4	74.1	6.4	0.5	0.06
347.50	77.9	14.8	99.8	8.7	0.6	0.10
347.55	46.1	8.9	39.0	3.4	0.8	0.04
347.63			K/T boundary			
347.70	64.9	12.5	57.1	5.0	0.8	0.05
347.80	72.0	14.1	37.3	3.2	1.4	0.04
347.90	56.5	11.1	23.2	2.0	1.7	0.11
347.95	89.2	17.6	33.4	2.9	1.9	0.04
348.15	128.0	17.7	74.9	4.6	1.3	0.15
348.35	222.7	30.8	113.4	6.9	1.4	0.17
348.40	157.0	30.7	86.6	7.5	1.3	0.09
348.50	128.0	24.7	109.3	9.5	0.8	0.10
349.00	114.0	22.2	67.7	5.9	1.2	0.10
349.50	75.0	14.6	49.5	4.3	1.1	0.06
349.90	60.0	11.8	26.1	2.3	1.6	0.05
350.00	72.0	14.1	36.9	3.2	1.4	0.07
350.50	81.0	15.7	55.8	4.8	1.0	0.09
351.06	114.0	22.2	67.2	5.8	1.2	0.11

Notes: Magnetostratigraphic, lithostratigaphic and biostratigraphic data from Gubbio have been reported previously (3, 4). The sampling interval was every ~ 0.05-0.10 m within ±1 m of the Cretaceous/Tertiary (K/T) boundary and every 0.50 m further away. Samples were pulverized with a masonry power drill at the outcrop site or powdered with a mortar and pestle in the laboratory. An entire strip of K/T clay was sampled from both Gubbio and Monte Conero. The clay was gently powdered with a mortar and pestle and the powder was thoroughly mixed. For all samples aliquots of 1 to 3 g were leached in 10% acetic acid to remove carbonate. The residue was termed non-carbonate fraction. The leaching does not remove either ^{3}He or ^{4}He (1). The decarbonated residue was transferred to tin foil cups. The samples were fused under vacuum at temperatures in excess of 1300 ^{o}C. Repeat extraction on the samples were performed frequently and in all cases were at blank levels of $<0.2 \times 10^{-9}$ cc STP for ^{4}He and $<1 \times 10^{-15}$ cc STP for ^{3}He. Gas handling and mass spectrometric techniques are described in (5), The 1σ variation on ~300 standards of similar size to the samples analyzed during this project was 0.5% for ^{4}He and 3.0% for ^{3}He.

The mean extraterrestrial ^{3}He concentration ($[^{3}He]_{Et}$) in the interval from the base of magnetochron 29R to the K/T boundary is $55 \pm 2.4 \times 10^{-15}$ cc STP g^{-1}. Data from (1) have been included for calculating the mean ^{3}He concentration. The uncertainty in the mean $[^{3}He]_{Et}$ was calculated from

$$\sigma_{mean} = \sqrt{\frac{\sum_{i=1}^{n} \sigma_i^2}{n^2}}$$

where, σ_i is the 1σ uncertainty in $[^{3}He]_{Et}$ in an individual sample and n is the total number of samples. The 1σ uncertainty on an individual sample is $20\% / \sqrt{N}$, where N is the number of measurements of the sample (1, 2).

The base of magnetochron 29R is at 65.58 Ma (7) and the age of the K/T boundary is ~ 65.0 Ma (8). Using a density of 2.7 g cm^{-3} for the Gubbio limestones (1), the average mass accumulation rate (MAR) in this interval is 1.9 g cm^{-2} kyr^{-1} and the average ^{3}He accretion rate (the product of $[^{3}He]_{Et}$ and MAR; 1, 6) is $106 \pm 4.6 \times 10^{-15}$ cc cm^{-2} kyr^{-1}. The uncertainty in the ^{3}He accretion rate is given by, $\sigma_{flux} = \sigma_{mean} \times MAR$.

The $[^3\text{He}]_{\text{Et}}$-based sedimentation rate in mm kyr^{-1}, s, was computed using the following relation:

$$s = \frac{f_{3He}\, r}{[^3\text{He}]_{Et} \times \rho}$$

The product $f_{3He}r$ is the average ^3He accretion rate ($1, 6$), where f_{3He} is the extraterrestrial ^3He accretion rate, r is a retentivity parameter that accommodates diagenetic and/or diffusional He losses, varying between unity and zero. $[^3\text{He}]_{\text{Et}}$ is the extraterrestrial ^3He concentration in the sediment, and ρ is the sediment density of 2.7 g cm^{-3} (*1*).

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