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# Pre-nuclear level of $^{129}\text{I}$ in Chinese loess-paleosol sections: A search for the natural $^{129}\text{I}$ level for dating in terrestrial environments

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**Abstract**

Due to its long half-life (15.7 Myr), radioactive  $^{129}\text{I}$  has great potential for dating geologic materials as old as 100 Myr. Thus, knowing the natural level of  $^{129}\text{I}$  is crucial to dating applications. The initial ratio of  $^{129}\text{I}/^{127}\text{I}$  in the ocean has been quantified by a number of researchers who have reached a consensus value. However, the applicability of  $^{129}\text{I}$  dating in the terrestrial environment remains problematic because the lack of an initial  $^{129}\text{I}/^{127}\text{I}$  value. In this work, samples of loess-paleosol sections from the Chinese Loess Plateau (CLP) were analyzed for  $^{129}\text{I}/^{127}\text{I}$ , aiming to provide an initial  $^{129}\text{I}/^{127}\text{I}$  ratio that can be adopted for dating purposes in terrestrial environments. A value of  $(2.0 \pm 1.0) \times 10^{-11}$  for the  $^{129}\text{I}/^{127}\text{I}$  ratio was found in two investigated loess-paleosol sections from Xifeng and Luochuan, China. This ratio is one order of magnitude higher than the initial value reported for the marine environment. Alteration of the natural initial  $^{129}\text{I}$  in the investigated samples due to the downward migration of anthropogenic  $^{129}\text{I}$  and by excess fissiogenic  $^{129}\text{I}$  from uranium was not supported. Consequently, the  $^{129}\text{I}/^{127}\text{I}$  ratio measured is considered to be a pristine value, and the difference from that in the marine systems is attributed to an isotopic dilution effect.

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**Keywords:**  $^{129}\text{I}$ ; Dating; Initial  $^{129}\text{I}/^{127}\text{I}$  ratio; Loess; AMS**1. INTRODUCTION**

Dating of geological materials using radioactive isotopes plays an important role in the understanding of the Earth's temporal evolution. However, dating methods available for sedimentary rocks has always been challenging, due to the lack of either a suitable radioactive isotope or materials for dating. Exploring and establishing a new geochronometer, especially for absolute dating, will without doubt expand and advance research in the Earth Sciences. An

excellent candidate is the radioactive isotope  $^{129}\text{I}$ , that has a half-life of 15.7 Myr and is continuously provided to the geological environment through natural input.  $^{129}\text{I}$  is naturally produced via cosmic rays interaction with xenon (Xe) in the upper atmosphere, spontaneous fission of  $^{238}\text{U}$ , thermal neutron-induced fission of  $^{235}\text{U}$  and to a lesser extent neutron activation reactions of tellurium, e.g.  $^{128}\text{Te}(n, \gamma)^{129}\text{Te}(\beta^-)^{129}\text{I}$  and  $^{130}\text{Te}(n, 2n)^{129}\text{Te}(\beta^-)^{129}\text{I}$ , in the Earth's crust.

Despite the early inference of Edwards (1962), who suggested that radioactive  $^{129}\text{I}$  was a potentially useful dating tool, the determination of the initial natural concentration of the isotope in geological archives has remained elusive, and a topic of  $^{129}\text{I}$  research for several decades. Due to

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the long residence time of  $^{129}\text{I}$  in ocean reservoirs ( $\sim 40$  kyr), much longer than the turnover time of sea water ( $\sim 1$  kyr),  $^{129}\text{I}$  was thought to be in equilibrium with stable iodine ( $^{127}\text{I}$ ). The ocean serves as an effective buffer against fluctuations in natural processes in the pre-nuclear era, i.e. before 1940 (Fabryka-Martin et al., 1985), so early research focused on the initial value in the marine environment. Because of the limitation in the measurement techniques, samples with an  $^{129}\text{I}/^{127}\text{I}$  ratio  $< 10^{-10}$  could not be measured until the early 1980s. Some estimates of an  $^{129}\text{I}/^{127}\text{I}$  ratio of  $(3\text{--}4) \times 10^{-12}$  and  $(5.5\text{--}6.5) \times 10^{-13}$  were reported using theoretical approaches (Fabryka-Martin et al., 1985). With the development of the accelerator mass spectrometry (AMS) technique for the measurement of  $^{129}\text{I}$  (Elmore et al., 1980), an average  $^{129}\text{I}/^{127}\text{I}$  ratio of  $1.2 \times 10^{-12}$  was obtained from marine sediments from Cape Hatteras in the Atlantic Ocean (Fehn et al., 1986). These data, together with other measurements of sediments collected along the east Pacific coast, provided an initial  $^{129}\text{I}/^{127}\text{I}$  value of  $1.5 \times 10^{-12}$  (Moran et al., 1998). A number of studies that dated geological samples formed in the marine environment have used this initial value. Examples include dating of pore waters associated with gas hydrates (Fehn et al., 2000; Lu et al., 2008; Tomaru et al., 2007) and the recycling of iodine in subduction zones (Snyder and Fehn, 2002). However, corresponding research using  $^{129}\text{I}/^{127}\text{I}$  in terrestrial environments was less successful because of the large scatter of initial values from terrestrial reservoirs. Pre-nuclear era samples were analyzed for  $^{129}\text{I}/^{127}\text{I}$  in Russian soil and in animal thyroid collected in the USA, and gave  $^{129}\text{I}/^{127}\text{I}$  values of  $10^{-12}$ – $10^{-11}$  (Sizdat et al., 2000). Higher values of  $2 \times 10^{-11}$  and  $(4\text{--}7) \times 10^{-10}$  were reported as pre-nuclear levels in river sediments from Spain and lake sediments from Sweden, respectively (Englund et al., 2008; Santos et al., 2007). These high ratios were attributed to anthropogenic  $^{129}\text{I}$  input. Jabbar et al. (2013) analyzed petrified wood samples collected in Austria and reported  $^{129}\text{I}/^{127}\text{I}$  ratios of  $1.15 \times 10^{-12}$  and  $0.63 \times 10^{-12}$ . Checking with the sedimentation age of corresponding strata, the initial value applied for the marine system  $1.5 \times 10^{-12}$  fit well for these samples. However, just one attempt is insufficient to confirm the applicability of this approach. As described above, there are few data and no agreement between results with respect to the initial natural  $^{129}\text{I}/^{127}\text{I}$  in the terrestrial environment.

Since the 1940s, human nuclear activities (e.g. nuclear weapon tests, discharge by nuclear fuels reprocessing plants and nuclear accidents) have released large amounts of  $^{129}\text{I}$  to the environment which has overwhelmed the natural environmental level (Fan et al., 2013; Hou et al., 2009). Thus, finding suitable samples that are as much as possible hidden from anthropogenic contamination is a challenging endeavor. We have selected sediment samples from the eolian deposits of the Chinese Loess Plateau (CLP) for the evaluation of the initial natural  $^{129}\text{I}$  concentration. These terrestrial sediments are composed of loess and paleosols,  $> 100$  m thick, first deposited about 2.6 Myr ago, and presenting a natural and continuous accumulation processes (An et al., 1991; Ding et al., 2005; Guo et al., 2002). Therefore, these materials are a suitable candidate

for studying the natural isotopic composition of iodine in the terrestrial environment. Earlier analysis of samples collected from depths of 23.5 and 63.5 m indicated  $^{129}\text{I}/^{127}\text{I}$  ratios of  $(1.6\text{--}1.8) \times 10^{-11}$  (Hou et al., 2010). Analysis of  $^{129}\text{I}$  speciation in some loess samples provided an  $^{129}\text{I}/^{127}\text{I}$  ratios of  $(1\text{--}2) \times 10^{-11}$  at depths of 90 cm and 60 cm in two loess sections from Sanyimiao and Luochuan, respectively (Luo et al., 2013). These results suggest a difference in the  $^{129}\text{I}/^{127}\text{I}$  values between the loess and marine sediments. In the work presented here, we have used two long loess-paleosol sections in order to provide a deeper insight into the pre-nuclear  $^{129}\text{I}$  level in the Chinese loess and to better constrain the initial value for  $^{129}\text{I}/^{127}\text{I}$  ratio and the possibility for dating.

## 2. MATERIALS AND METHODS

Samples used for the depth distribution of  $^{129}\text{I}$  were collected in loess-paleosol sections from two localities: Xifeng ( $35.79^\circ\text{N}$ ,  $107.59^\circ\text{E}$ ) and Luochuan ( $35.78^\circ\text{N}$ ,  $109.44^\circ\text{E}$ ) (Fig. 1a). The localities occur in the middle part of the CLP and the sections have been studied as paleoclimate archives (An et al., 1991; Heller and Liu, 1982; Kukla and An, 1989; Liu, 1985; Sun et al., 2006; Sun et al., 2015; Zhu et al., 2007). The Xifeng section, 73.7 m deep, was sampled in 2007. The bottom reached loess unit  $L_9$ , corresponding to an age of around 900 kyr (Sun et al., 2006). The Luochuan section was sampled in 2004 with a maximum depth of 12.45 m, reaching the bottom of paleosol unit  $S_1$ , which corresponds to 130 kyr (Zhou et al., 2010). A trough was excavated to remove the overlying soil in order to avoid possible contamination from modern  $^{129}\text{I}$  before sampling in both sites (Fig. 1b). Samples were taken in small intervals (10–20 cm) in the upper parts of the Xifeng section while larger intervals (0.5–5 m) were obtained for the lower parts. For each sample, a thickness of 1–2 cm was taken, and the top of each segment was recorded as the depth of the corresponding sample.

All the loess-paleosol samples were air-dried, ground and sieved through a 200-mesh sieve. 20.0 g of each sample was taken for the analysis of  $^{129}\text{I}$  and  $^{127}\text{I}$ . According to our previous study, the loess and paleosols were found to be low in both  $^{127}\text{I}$  ( $\sim 2 \mu\text{g/g}$ ) and  $^{129}\text{I}$  ( $\sim 10^5$  atoms/g) (Hou et al., 2010). Therefore, the carrier free iodine separation method was adopted for the extraction of iodine from all samples in order to avoid any possible contamination induced by carrier iodine.  $^{129}\text{I}/^{127}\text{I}$  atomic ratios in the prepared targets were measured by accelerator mass spectrometry using the 3 MV Tandem AMS system at the Xi'an AMS center. The details of the extraction procedure and AMS measurements followed the description by Hou et al. (2010).

Seven blank samples in total, undergoing the same procedure as the loess samples but without soils, were added, to monitor the chemical background during processing of the loess samples. Because of the significant difference of iodine content in the targets between samples and blanks due to the utility of carrier free iodine method, the  $^{129}\text{I}$  count rate and the  $^{127}\text{I}$  beam intensity were taken instead of  $^{129}\text{I}/^{127}\text{I}$  ratio for description of the measurement of the blanks. The  $^{129}\text{I}$  count rate in the loess samples ( $> 25$

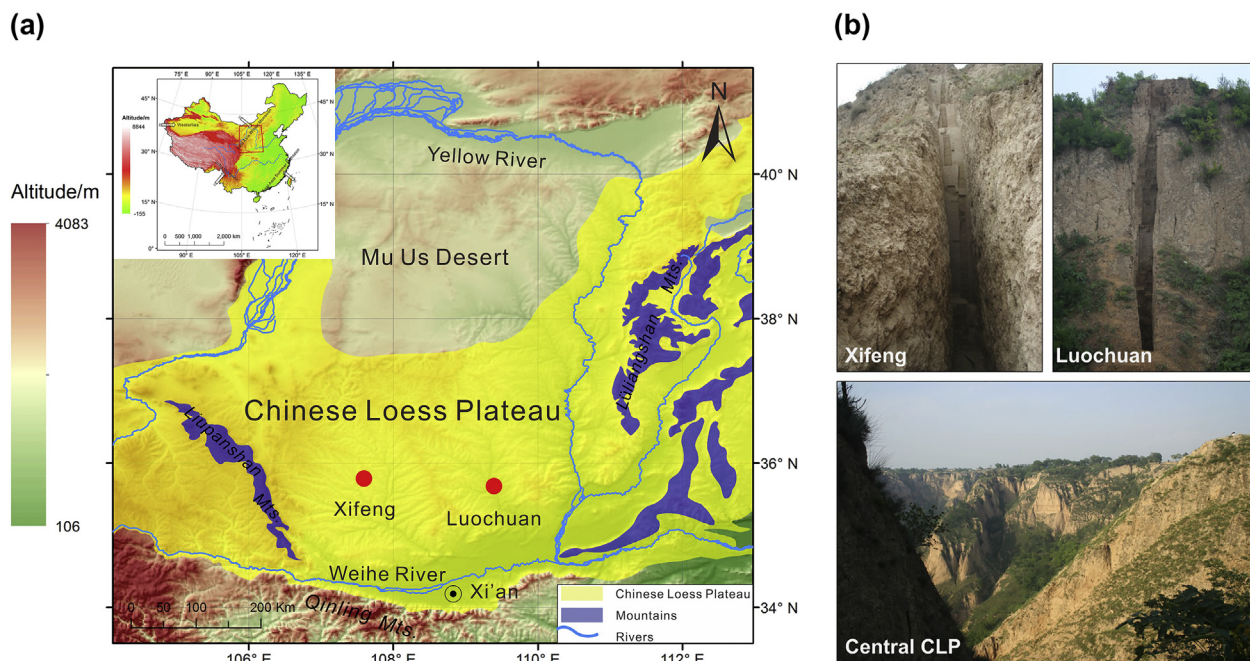


Fig. 1. Location (a) and outcrop scene (b) of the sampling sites. The bright yellow area in (a) represents the extent of the CLP. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

counts/min) was  $>5$  times that in the blank samples ( $<5$  counts/min), while  $^{127}\text{I}$  beam intensity in 5 min in the loess samples ( $>5 \times 10^{-7}$  coulomb) was  $>6$  times that in the blank samples ( $<8 \times 10^{-8}$  coulomb). Therefore, the significant contamination of  $^{129}\text{I}$  and  $^{127}\text{I}$  during the chemical analysis can be excluded.

Iodine ( $^{127}\text{I}$ ) was measured by ICP-MS using an X SeriesII ICP-MS (Thermal Electron Corporation), under hot plasma conditions, with an Xt interface. Iodine extracted from soil samples by combustion at  $800\text{ }^\circ\text{C}$  was trapped in a  $\text{NaOH-NaHSO}_3$  solution, which was diluted 10 times using deionized water ( $18.2\text{ M}\Omega$ ), and a  $\text{Cs}^+$  solution was added to the samples, blanks and standards to a final concentration of 2.0 ppb as an internal standard. The detection limit of this method for iodine is estimated as 3 times blank to be 0.02 ppb. An  $0.15\text{ mol/L NH}_3\cdot\text{H}_2\text{O}$  solution was used to rinse the samples during the ICP-MS measurements. The  $^{129}\text{I}$  concentration was calculated using the measured  $^{129}\text{I}/^{127}\text{I}$  atomic ratio by AMS and the  $^{127}\text{I}$  concentration by ICP-MS.

For each sample, a  $0.2000\text{ g}$  subsample was taken for uranium analysis. The samples were first ashed in an oven at  $550\text{ }^\circ\text{C}$  for 3 h, and then digested with  $\text{HNO}_3$  and  $\text{HF}$  in a Teflon crucible, and then evaporated on a hot plate. The residues were dissolved in 3%  $\text{HNO}_3$  for the ICP-MS measurement, in which the  $\text{In}^{3+}$  solution was added and used as an internal standard.

### 3. RESULTS

Table 1 lists the analytical results of iodine and uranium isotopes in the two loess-paleosol sections. The  $^{129}\text{I}/^{127}\text{I}$  atomic ratios for the Xifeng and Luochuan loess sections

are  $(0.65\text{--}16.30) \times 10^{-11}$  and  $(0.39\text{--}484.72) \times 10^{-11}$ , respectively. The highest values of the  $^{129}\text{I}/^{127}\text{I}$  ratio as well as the  $^{129}\text{I}$  concentration occur in the surface layer in both sections. These are comparable with the level in surface soil from the Guanzhong area,  $(1.08\text{--}28.61) \times 10^{-10}$  (Zhang et al., 2011), about 200 km south of Luochuan, indicating that the  $^{129}\text{I}$  in the surface environment in the CLP has the same anthropogenic  $^{129}\text{I}$  sources as that for the Guanzhong area, which was dominated by atmospheric nuclear weapon tests and discharge from nuclear fuel reprocessing plants. It is worth mentioning that the  $^{129}\text{I}$  signals in the top parts of the Xifeng section ( $1.6 \times 10^{-10}$ ) were much lower than those from the Luochuan section ( $4.8 \times 10^{-9}$ ). This might be attributed to missing top soils from Xifeng before sampling. There is evidence for this, as it was reported that the black marker layer  $\text{S}_0$ , that formed during a warm-humid climate after the last glacial period, was found to be missing in field studies at the Xifeng site (Porter and An, 1995). Soils from the loess plateau were widely used for construction by the local inhabitants. This could explain the missing surface layers. In addition, loess profile sampling by excavation was usually carried out at the edge of a loess tableland. Collapse near the tableland edge could also cause the uppermost sediments to be lost. However, the top strata in the Luochuan site were better preserved, with an intact  $\text{S}_0$  horizon. The  $^{129}\text{I}/^{127}\text{I}$  for the two sections decrease with increasing depth, and then maintain relatively constant values around  $2.0 \times 10^{-11}$  from 1.6 m and 0.6 m, in the Xifeng and Luochuan sections, respectively (Fig. 2). The depth distributions of  $^{129}\text{I}$  concentrations in both sections show variation comparable to the  $^{129}\text{I}/^{127}\text{I}$  trend, while the  $^{127}\text{I}$  concentration does not show any apparent trend with depth.

Table 1

Analytical results of iodine and uranium isotopes in the Xifeng and Luochuan loess-paleosol sections. <sup>a</sup>An age correction was made on the <sup>129</sup>I/<sup>127</sup>I ratios and <sup>129</sup>I concentrations of samples deeper than 1.5 m and 0.4 m from the Xifeng and Luochuan sections, respectively, based on the ages of bottom layers in both loess sections and the assumption of a roughly constant sedimentation rate; <sup>b</sup>The <sup>129</sup>I/<sup>127</sup>I ratios of samples at depths of 23.5 m and 63.5 m from the Xifeng section were cited from previous work by Hou et al. (2010) and were also age-corrected; NM: not measured.

Loess section	Sample No.	Depth m	<sup>129</sup> I/ <sup>127</sup> I atom ratio <sup>a</sup> × 10 <sup>-11</sup>	<sup>129</sup> I concentration <sup>a</sup> × 10 <sup>5</sup> atoms/g	<sup>127</sup> I concentration μg/g	U concentration μg/g	<sup>235</sup> U/ <sup>238</sup> U ratio × 10 <sup>-3</sup>
<i>Xifeng</i>							
	XF07-20	0.2	16.12 ± 0.64	13.70 ± 0.58	1.83 ± 0.03	1.929 ± 0.039	7.310 ± 0.222
	XF07-30	0.3	6.06 ± 0.22	4.54 ± 0.23	1.61 ± 0.06	1.693 ± 0.032	7.738 ± 0.209
	XF07-40	0.4	16.30 ± 1.12	14.01 ± 1.20	1.85 ± 0.10	1.644 ± 0.031	7.352 ± 0.208
	XF07-50	0.5	4.75 ± 0.53	4.04 ± 0.49	1.82 ± 0.09	1.852 ± 0.034	7.616 ± 0.202
	XF07-60	0.6	5.22 ± 0.38	4.50 ± 0.45	1.77 ± 0.12	1.645 ± 0.031	7.348 ± 0.203
	XF07-80	0.8	3.19 ± 0.24	2.64 ± 0.21	1.78 ± 0.05	1.566 ± 0.031	7.722 ± 0.217
	XF07-100	1.0	2.76 ± 0.29	2.34 ± 0.25	1.82 ± 0.04	1.241 ± 0.023	7.305 ± 0.202
	XF07-140	1.4	5.47 ± 0.15	4.79 ± 0.33	1.88 ± 0.12	1.639 ± 0.032	7.375 ± 0.207
	XF07-150	1.5	5.45 ± 0.71	5.85 ± 0.77	2.39 ± 0.06	1.460 ± 0.027	7.591 ± 0.219
	XF07-160	1.6	0.65 ± 0.04	0.81 ± 0.06	2.64 ± 0.12	1.705 ± 0.033	7.683 ± 0.216
	XF07-200	2.0	2.25 ± 0.27	2.61 ± 0.31	2.45 ± 0.05	1.713 ± 0.033	7.647 ± 0.211
	XF07-250	2.5	1.80 ± 0.25	2.11 ± 0.29	2.48 ± 0.06	1.704 ± 0.031	7.687 ± 0.209
	XF07-300	3.0	2.10 ± 0.19	1.94 ± 0.20	1.95 ± 0.10	1.835 ± 0.035	7.688 ± 0.215
	XF07-350	3.5	2.09 ± 0.24	2.26 ± 0.26	2.28 ± 0.06	2.029 ± 0.037	7.447 ± 0.211
	XF07-400	4.0	2.28 ± 0.22	2.40 ± 0.27	2.22 ± 0.14	2.046 ± 0.036	7.385 ± 0.206
	XF07-450	4.5	1.96 ± 0.20	1.96 ± 0.24	2.11 ± 0.16	2.161 ± 0.040	7.459 ± 0.206
	XF07-500	5.0	1.19 ± 0.08	1.11 ± 0.08	1.98 ± 0.06	2.053 ± 0.037	7.360 ± 0.199
	XF07-600	6.0	0.70 ± 0.05	2.38 ± 0.20	7.17 ± 0.35	NM	NM
	XF07-700	7.0	0.67 ± 0.06	1.63 ± 0.14	5.13 ± 0.08	NM	NM
	XF07-850	8.5	2.58 ± 0.26	2.31 ± 0.26	1.89 ± 0.10	2.083 ± 0.038	7.253 ± 0.193
	XF07-1000	10.0	0.81 ± 0.06	2.85 ± 0.19	7.39 ± 0.20	NM	NM
	XF07-1340	13.4	7.90 ± 0.73	3.59 ± 0.36	0.89 ± 0.03	1.948 ± 0.036	7.759 ± 0.204
	XF07-1850	18.5	2.71 ± 0.28	2.35 ± 0.25	1.83 ± 0.05	1.855 ± 0.035	7.604 ± 0.212
	XF07-2350	23.5	1.69 ± 0.53 <sup>b</sup>	1.99 ± 0.62	2.49 ± 0.02	1.783 ± 0.033	7.344 ± 0.203
	XF07-2850	28.5	3.33 ± 0.32	2.24 ± 0.23	1.42 ± 0.07	1.627 ± 0.031	7.430 ± 0.211
	XF07-3350	33.5	2.23 ± 0.30	1.10 ± 0.15	1.05 ± 0.05	1.741 ± 0.031	7.523 ± 0.195
	XF07-3850	38.5	3.70 ± 0.32	1.84 ± 0.18	1.05 ± 0.06	1.747 ± 0.032	7.497 ± 0.209
	XF07-4350	43.5	2.25 ± 0.22	1.70 ± 0.16	1.60 ± 0.01	1.521 ± 0.028	7.284 ± 0.200
	XF07-2-500	48.5	2.90 ± 0.32	1.55 ± 0.17	1.13 ± 0.04	1.710 ± 0.031	7.660 ± 0.211
	XF07-2-1000	53.5	3.95 ± 0.38	2.30 ± 0.25	1.23 ± 0.06	1.845 ± 0.033	7.646 ± 0.210
	XF07-2-1500	58.5	2.31 ± 0.21	2.36 ± 0.23	2.16 ± 0.06	1.689 ± 0.030	7.756 ± 0.202
	XF07-2-2000	63.5	1.89 ± 0.42 <sup>b</sup>	2.26 ± 0.55	2.52 ± 0.24	1.473 ± 0.028	7.523 ± 0.207
	XF07-2-2500	68.5	3.87 ± 0.42	2.31 ± 0.26	1.26 ± 0.05	1.550 ± 0.029	7.802 ± 0.205
	XF07-2-3000	73.5	3.27 ± 0.35	2.08 ± 0.27	1.34 ± 0.10	1.674 ± 0.031	7.220 ± 0.206
<i>Luochuan</i>							
	LC04-00	0.00	484.72 ± 10.98	429.66 ± 9.73	1.87 ± 0.03	0.692 ± 0.202	7.278 ± 0.202
	LC04-10	0.10	412.85 ± 14.79	420.75 ± 15.08	2.15 ± 0.07	0.962 ± 0.215	7.329 ± 0.215
	LC04-20	0.20	290.18 ± 22.43	290.23 ± 22.43	2.11 ± 0.16	0.859 ± 0.213	8.215 ± 0.213

LC04-30	0.30	121.64 ± 2.75	125.70 ± 2.84	2.18 ± 0.03	0.856 ± 0.213	8.244 ± 0.213
LC04-40	0.40	5.29 ± 0.25	5.44 ± 0.33	2.17 ± 0.09	0.902 ± 0.216	7.821 ± 0.216
LC04-60	0.60	1.52 ± 0.09	1.28 ± 0.09	1.78 ± 0.07	1.023 ± 0.211	7.881 ± 0.211
LC04-80	0.80	1.49 ± 0.04	1.50 ± 0.05	2.13 ± 0.03	1.012 ± 0.212	7.968 ± 0.212
LC04-100	1.00	0.62 ± 0.02	1.07 ± 0.10	3.67 ± 0.30	1.040 ± 0.210	7.751 ± 0.210
LC04-125	1.25	0.39 ± 0.04	1.86 ± 0.17	10.12 ± 0.42	NM	NM
LC04-175	1.75	1.50 ± 0.08	5.14 ± 0.55	7.23 ± 0.69	NM	NM
LC04-205	2.05	0.50 ± 0.03	0.50 ± 0.03	2.14 ± 0.13	1.408 ± 0.203	7.874 ± 0.203
LC04-215	2.15	2.82 ± 0.12	7.78 ± 0.86	5.82 ± 0.60	NM	NM
LC04-235	2.35	1.59 ± 0.10	2.47 ± 0.33	3.28 ± 0.40	NM	NM
LC04-255	2.55	2.19 ± 0.06	1.41 ± 0.16	1.36 ± 0.15	1.601 ± 0.202	7.551 ± 0.202
LC04-495	4.95	6.16 ± 0.16	6.22 ± 0.23	2.13 ± 0.06	1.340 ± 0.141	7.518 ± 0.141
LC04-755	7.55	3.61 ± 0.13	2.68 ± 0.15	1.57 ± 0.07	1.543 ± 0.143	7.838 ± 0.143
LC04-915	9.15	1.60 ± 0.11	3.70 ± 0.28	4.87 ± 0.19	NM	NM
LC04-1005	10.05	2.05 ± 0.05	4.19 ± 0.11	4.30 ± 0.07	1.651 ± 0.138	7.321 ± 0.138
LC04-1100	11.00	1.03 ± 0.07	2.69 ± 0.22	5.49 ± 0.06	NM	NM
LC04-1205	12.05	2.62 ± 0.12	1.69 ± 0.12	1.36 ± 0.08	1.469 ± 0.144	7.544 ± 0.144
LC04-1245	12.45	2.09 ± 0.11	2.36 ± 0.17	2.38 ± 0.13	NM	NM

The concentrations of stable iodine in the Xifeng section varied from 0.89 to 7.39  $\mu\text{g/g}$  with an average of 2.25  $\mu\text{g/g}$ , and that in the Luochuan section from 1.36 to 10.12  $\mu\text{g/g}$  with an average of 3.34  $\mu\text{g/g}$ . These values are comparable with the reported iodine content in soil (1–3  $\mu\text{g/g}$ ), with a few exceptions, but much lower than those in marine sediments (generally 20–200  $\mu\text{g/g}$ ) (Hu and Moran, 2010). The relatively high  $^{127}\text{I}$  concentrations of 5–7  $\mu\text{g/g}$  at depths of 6–10 m in the Xifeng section and 7–10  $\mu\text{g/g}$  at 1–2 m in the Luochuan section were observed and corresponded to the paleosol layers, which are rich in organic matter compared to the loess layers (Sun et al., 2011). This high  $^{127}\text{I}$  concentrations linked to the high organic matter content is consistent with the dominant organic fraction of iodine observed in the loess soils (Luo et al., 2013).

The uranium concentration was measured to be 1.241–2.161  $\mu\text{g/g}$  in the Xifeng section and slightly lower in the Luochuan section, 0.692–1.651  $\mu\text{g/g}$ . The  $^{235}\text{U}/^{238}\text{U}$  ratio varied within  $(7.22\text{--}8.24) \times 10^{-3}$ . These data are comparable with the uranium concentration in soils from Europe (0.8–11.0  $\mu\text{g/g}$ ) and the United States (2.3–3.7  $\mu\text{g/g}$ ) and with the natural composition of  $^{235}\text{U}/^{238}\text{U}$   $7.250 \times 10^{-3}$  (Kabatapendias and Pendias, 1984; Lella et al., 2005).

#### 4. DISCUSSION

It was reported that the accumulation rates of the loess and paleosol in the Xifeng and Luochuan sections are 3.8–16.5  $\text{cm/kyr}$  (Wu et al., 1998). Thus, layers deeper than 4 cm were formed approximately 1000 years ago, much earlier than human nuclear activities (1940s). If no vertical migration of iodine occurred, the  $^{129}\text{I}/^{127}\text{I}$  in the layers below 4 cm should represent the natural level of  $^{129}\text{I}$  in the loess-paleosol sections. However, the  $^{129}\text{I}/^{127}\text{I}$  ratio at a depth of about 10 cm is almost equal to that in the surface layer, and the ratio remains relatively high down to a depth of 40–60 cm (Fig. 2). This feature indicates vertically downward migration of anthropogenic  $^{129}\text{I}$  to some extent. In layers deeper than 2 m, much lower and relatively constant  $^{129}\text{I}/^{127}\text{I}$  ratios were observed in the two soil sections. The average ratio and one standard deviation of  $^{129}\text{I}/^{127}\text{I}$  ratios in the deep layers ( $>2$  m) in the two sections is calculated to be  $(2.0 \pm 1.0) \times 10^{-11}$  excepting two relatively high values of  $7.9 \times 10^{-11}$  from a layer at 13.4 m in the Xifeng section and  $6.2 \times 10^{-11}$  at 4.95 m in the Luochuan section, which might represent the natural level of  $^{129}\text{I}$  in the loess and paleosols. It is not clear why a slightly high  $^{129}\text{I}/^{127}\text{I}$  value of  $(5\text{--}10) \times 10^{-11}$  occurred in two layers in the two loess sections, a further investigation on variation of the iodine isotopes in this region is planned. This  $^{129}\text{I}/^{127}\text{I}$  value,  $(2.0 \pm 1.0) \times 10^{-11}$ , is about one order of magnitude higher than that observed in the deep marine sediments,  $(1.5 \pm 0.15) \times 10^{-12}$  (Moran et al., 1998). This difference can probably be caused by: (1) severe downward migration of anthropogenic  $^{129}\text{I}$  from the surface layers (reaching tens of meters), (2) a significant contribution of uranium fission product, and (3) disequilibrium of iodine isotopes between the marine and terrestrial environments and therefore their different pre-nuclear  $^{129}\text{I}/^{127}\text{I}$  ratios. Below we discuss the influence of each of these processes.

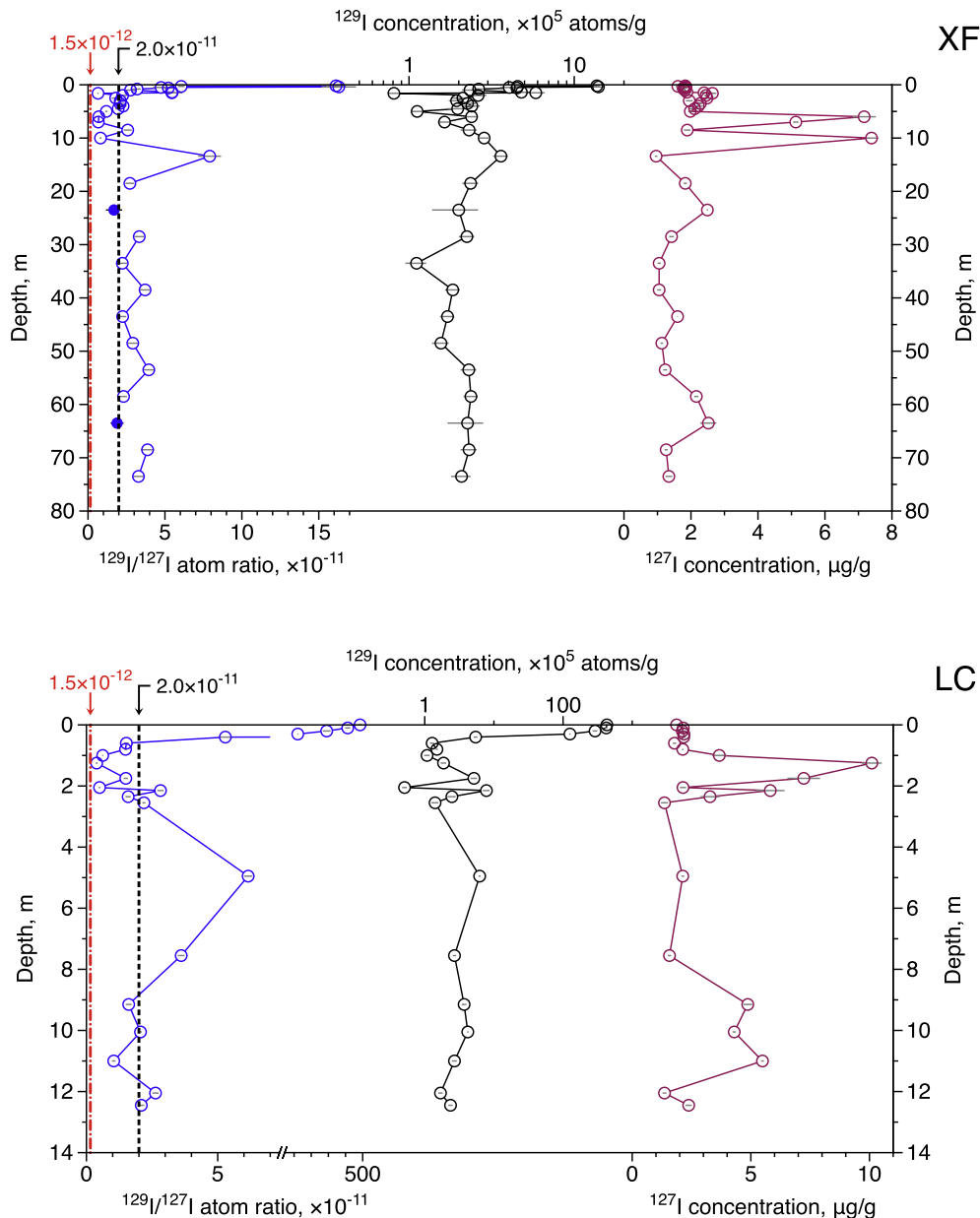


Fig. 2. Depth distribution of  $^{129}\text{I}/^{127}\text{I}$  and concentrations of  $^{129}\text{I}$  and  $^{127}\text{I}$  in the loess-paleosol sections of Xifeng (XF) and Luochuan (LC). Solid blue circle:  $^{129}\text{I}/^{127}\text{I}$  ratio at 23.5 m and 63.5 m depth in the XF section are from Hou et al. (2010). The black dashed line represents the pre-nuclear  $^{129}\text{I}/^{127}\text{I}$  of  $2.0 \times 10^{-11}$  in the CLP and the red dot-dashed line represents the initial  $^{129}\text{I}/^{127}\text{I}$  of  $1.5 \times 10^{-12}$  for the marine system. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

#### 4.1. Downward migration of anthropogenic $^{129}\text{I}$ in the loess-paleosol sections

The trend of  $^{129}\text{I}/^{127}\text{I}$  depth distribution supports limited downward migration of the anthropogenic  $^{129}\text{I}$  as a result of seepage driven by gravity flow of rainwater, and it is observed only at the top parts of the sections. The  $^{129}\text{I}/^{127}\text{I}$  values stabilize at a depth  $> 2$  m (Fig. 2) without showing a gradual decrease through much thicker layers. In laboratory experiments, the migration rate of iodine in the soil was investigated to vary from  $< 10^{-8}$  to  $10^{-7}$  cm/s

(Tikhomirov et al., 1981). Given that the  $^{129}\text{I}$  released from human nuclear activities since 1945 is deposited in the top layers of the Xifeng and Luochuan sections, and migrated into the deeper layers at a rate of  $10^{-7}$  cm/s, the downward migration distance from the surface can be estimated to be no more than 2 m until 2007, which is the sampling date. However, considering that the migration rate becomes sluggish due to transformation of soluble iodine into stable species strongly associated with soil particles (Tikhomirov et al., 1981), then the migration distance should be even shorter than 2 m. It has been reported that different

fractions/species of  $^{129}\text{I}$  exponentially decreased with increasing depth in loess-paleosol sections collected from the Sanyimiao and Luochuan sites (Luo et al., 2013). In all the fractions, including leachable, organic, oxides and residue,  $^{129}\text{I}$  concentrations decreased 100 times at 0.9 m and 0.5 m compared to the surface layers, respectively, indicating limited migration distance of iodine in the loess. The results obtained here also support limited depth-related migration of anthropogenic  $^{129}\text{I}$  which can be a maximum depth of 1.6 m and 0.6 m in the Xifeng and Luochuan sections, respectively.

#### 4.2. Contribution of uranium fissiogenic $^{129}\text{I}$

Uranium is an important source of natural  $^{129}\text{I}$  in the Earth's crust through spontaneous fission of  $^{238}\text{U}$  and neutron induced fission of  $^{235}\text{U}$ . These uranium fissiogenic processes might be a possible source of  $^{129}\text{I}$  in the loess and paleosols. Based on the uranium concentration (Table 1), the contribution of uranium-sourced fissiogenic  $^{129}\text{I}$  to the total  $^{129}\text{I}$  in the loess-paleosol samples can be estimated using the following equation (Boaretto et al., 1990):

$$^{129}\text{I} = (^{238}\text{U} \cdot f_s^{129} \cdot \lambda^{238} + ^{235}\text{U} \cdot f_i^{129} \cdot \varphi\sigma) \cdot (1 - e^{-\lambda^{129}t}) / \lambda^{129}, \quad (1)$$

where  $^{238}\text{U}$  and  $^{235}\text{U}$  are the measured concentrations ( $\mu\text{g/g}$ );  $f_s$  and  $f_i$  are the spontaneous fission yields of  $^{238}\text{U}$  (0.03%) and neutron-induced fission yield of  $^{235}\text{U}$  (0.75%), respectively;  $\lambda^{129}$  is the decay constant of  $^{129}\text{I}$  ( $4.4 \times 10^{-8} \text{ yr}^{-1}$ );  $\lambda^{238}$  is the  $^{238}\text{U}$  decay constant ( $8.5 \times 10^{-17} \text{ yr}^{-1}$ );  $\varphi\sigma$  is the production rate from thermal- and epithermal-neutron-induced fission of  $^{235}\text{U}$ ; and  $t$  is the residence time of the U phase in the material. Because the spontaneous fission production mode is expected to be dominant at low U concentrations ( $<100 \mu\text{g/g}$ ) (Boaretto et al., 1990), the equation can be re-written as:

$$^{129}\text{I} = ^{238}\text{U} \cdot f_s^{129} \cdot \lambda^{238} \cdot (1 - e^{-\lambda^{129}t}) / \lambda^{129} \quad (2)$$

The variable  $t$  refers to the onset of fissiogenic  $^{129}\text{I}$  by uranium in the samples. In the loess and paleosol samples, some uranium existed in the crystal lattice when the minerals formed (hundreds of millions of years), while some entered the soil from the atmosphere during pedogenesis (less than hundreds of thousands of years) after the eolian dust deposited onto the CLP. Since the variable  $^{129}\text{I}$  in the equation correlated positively with the independent variable  $t$ , a larger  $t$ , the age of the provenance of the loess and paleosol, is reasonable to provide an upper limit of the fissiogenic contribution. By means of detrital-zircon U-Pb dating, the dominant age populations in the Xifeng and Luochuan sites are found to be both roughly in the range of 200–500 Myr (Xiao et al., 2012). Supposing the uranium in the samples formed only within the minerals, and taking 500 Myr as the maximum age of the loess-paleosol minerals, a fissiogenic  $^{129}\text{I}$  content can be calculated to be around 100 atoms/g. This is  $\leq 0.1\%$  of the total  $^{129}\text{I}$  ( $10^5$ – $10^6$  atoms/g) in the samples in all the loess-paleosol strata studied. Therefore, the contribution of the fissiogenic  $^{129}\text{I}$  from uranium in the samples is negligible.

#### 4.3. Comparison of the pre-nuclear $^{129}\text{I}/^{127}\text{I}$ in loess with the marine system

The pre-nuclear  $^{129}\text{I}/^{127}\text{I}$  of  $2.0 \times 10^{-11}$  in the deep loess-paleosol sections of Xifeng and Luochuan is one order of magnitude higher than the value ( $1.5 \times 10^{-12}$ ) reported in the marine system. This difference might be attributed to the possible different isotopic equilibrium of iodine in the terrestrial and marine systems. The ocean is the major reservoir of  $^{127}\text{I}$  on the Earth's surface while the concentration of the radioactive  $^{129}\text{I}$  strongly depends on different production modes (cosmogenic and fissiogenic). Assuming a constant production of radioactive iodine on land and in the ocean, the relatively large inventory of stable iodine in the marine environment may dilute its  $^{129}\text{I}/^{127}\text{I}$  ratio, as compared to the terrestrial value. This mechanism could lead to a lower  $^{129}\text{I}/^{127}\text{I}$  value in the oceans, as suggested by Edwards and Rey (1968). The terrestrial stable iodine content is orders of magnitude lower than that of the marine environment (Hu and Moran, 2010). The production rate of cosmogenic  $^{129}\text{I}$  is same for both reservoirs, and the fissiogenic  $^{129}\text{I}$  production rate is about tenfold higher in the terrestrial environment than in the marine environment (Edwards and Rey, 1968). Therefore, the difference of  $^{129}\text{I}/^{127}\text{I}$  between terrestrial and marine environment may be an order of magnitude or more. A similar distribution pattern has been found for natural chlorine in groundwater, where the lowest  $^{36}\text{Cl}/\text{Cl}$  occurred near the coast, and the ratio increased to a maximum in the inland region of the United States (Davis et al., 2003). That was also interpreted as incomplete mixing of chlorine isotopes, since a large fraction of chlorine in the continent is of marine origin while  $^{36}\text{Cl}$  is a cosmogenic radionuclide. Although there is no evidence, such a distribution pattern for natural iodine isotopes is quite possible for the terrestrial environment because of the similar origin of stable iodine and chlorine. Thus, the extent of iodine isotopic exchange between the two systems would be another important factor impacting the isotopic ratio for the terrestrial environment, especially for the environment of the CLP which is more than 1000 km from the Pacific Ocean. According to the assumption above, we tried to make a calculation of the  $^{129}\text{I}/^{127}\text{I}$  ratio in the terrestrial environment based on the valid inventory of the two isotopes corresponding to the land surface. The equilibrium contribution of natural  $^{129}\text{I}$  to the hydrosphere calculated by Edwards and Rey (1968) was adopted as the input of  $^{129}\text{I}$  to the terrestrial environment, in which the data for weathering of rocks, volcanic matter and cosmogenic production are  $3.6 \times 10^{25}$ ,  $2.3 \times 10^{26}$  and  $5.8 \times 10^{26}$  atoms, respectively. The input  $^{129}\text{I}$  was calculated to be  $4.34 \times 10^{26}$  atoms, considering a reasonable cosmogenic  $^{129}\text{I}$  input corresponding to the land area percentage of the Earth's surface (29%). As discussed above, referring to the  $^{129}\text{I}$  downward migration, the iodine in the loess/soil sections cannot penetrate more than 2 m downward, and we expect much less penetration in rocks, due their low porosity. Thus, we arbitrarily used 2 m as a global maximum thickness for all exposed rocks on the land surface, that are participating in the exchange of iodine isotopes with the atmosphere. Considering the surface area of the

continents ( $1.49 \times 10^8 \text{ km}^2$ ) (<https://en.wikipedia.org/wiki/Continent>), the average density of the continental crust ( $2.7 \text{ g/cm}^3$ ), the area percentage (73%) of sedimentary rocks (Wilkinson et al., 2009), the average iodine concentration for sedimentary rocks ( $1.49 \text{ }\mu\text{g/g}$ ) and for other rock types ( $0.024 \text{ }\mu\text{g/g}$ , for metamorphic rocks, the highest value of the upper crust being taken as the iodine concentration in all the other types except sedimentary rocks) (Muramatsu and Wedepohl, 1998) into account, the total inventory of  $^{127}\text{I}$  is calculated to be  $4.2 \times 10^{36}$  atoms. Then, the  $^{129}\text{I}/^{127}\text{I}$  is estimated at  $1.0 \times 10^{-10}$ . This value might represent an isotopic composition of terrestrial iodine (upper limit) under a non-exchange mode. When exchange is considered, more stable iodine than  $^{129}\text{I}$  would be transported from ocean to land. Therefore, terrestrial  $^{129}\text{I}/^{127}\text{I}$  depends on how much  $^{127}\text{I}$  is of marine origin. The Chinese loess-paleosol pre-anthropogenic  $^{129}\text{I}/^{127}\text{I}$  resides between the theoretically calculated terrestrial ratio  $1.0 \times 10^{-10}$  and the  $1.5 \times 10^{-12}$  of the marine system, indicating a contribution of marine  $^{127}\text{I}$  to the terrestrial environment. Consequently, the results of the two soil sections and the discussion above suggest that the measured  $^{129}\text{I}/^{127}\text{I}$  ratio of  $(2.0 \pm 1.0) \times 10^{-11}$  at the depth provides the best estimate of the initial  $^{129}\text{I}$  pre-nuclear natural level in the terrestrial system of the CLP and most likely in mid-latitude regions of the Northern Hemisphere.

## 5. CONCLUSIONS

The results and discussion presented here lead to the following conclusions: (1) A constant low  $^{129}\text{I}/^{127}\text{I}$  of about  $(2.0 \pm 1.0) \times 10^{-11}$  was observed in the deep layers of loess-paleosol sections from the Xifeng and Luochuan sites, China; (2) the pre-nuclear level of  $^{129}\text{I}/^{127}\text{I}$  in the loess deposits is one order of magnitude higher than the reported initial value  $(1.5 \pm 0.15) \times 10^{-12}$  derived from marine sediments; (3) possible anthropogenic  $^{129}\text{I}$  input via downward migration and excessive fissiogenic  $^{129}\text{I}$  from uranium isotopes have negligible impact on the iodine isotopic ratio. The average value of  $(2.0 \pm 1.0) \times 10^{-11}$  for the  $^{129}\text{I}/^{127}\text{I}$  provides a reasonable estimate of the initial natural  $^{129}\text{I}$  level in the terrestrial environment of the CLP and most likely in the middle latitudes of the Northern Hemisphere; (4) the higher pre-nuclear  $^{129}\text{I}/^{127}\text{I}$  value of the terrestrial environment compared to the marine system might be attributed to low stable  $^{127}\text{I}$  concentration in the terrestrial environment. These findings may open new frontiers of research utilizing  $^{129}\text{I}$  as a dating tool in the terrestrial system and provide a better constraint on the pre-nuclear  $^{129}\text{I}$  level in the terrestrial environment globally.

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