$^{135}\text{Cs}/^{137}\text{Cs}$ Isotopic Composition of Environmental Samples Across Europe: Environmental Transport and Source Term Emission Applications

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Abstract

$^{135}\text{Cs}/^{137}\text{Cs}$ isotopic analyses represent an important tool for studying the fate and transport of radiocesium in the environment; in this work the $^{135}\text{Cs}/^{137}\text{Cs}$ isotopic composition in environmental samples taken from across Europe is reported. Surface soil and vegetation samples from the western Russia, Ukraine, Austria, and Hungary show consistent aged thermal fission product $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios of 0.58 ± 0.01 (age corrected to 1/1/15), with the exception of one sample of soil-moss from Hungary which shows an elevated $^{135}\text{Cs}/^{137}\text{Cs}$ ratio of 1.78 ± 0.12 . With the exception of the outlier sample from Hungary, surface soil/vegetation data are in quantitative agreement with values previously reported for soils within the Chernobyl exclusion zone, suggesting that radiocesium at these locations is primarily composed of homogenous airborne deposition from Chernobyl. Seawater samples taken from the Irish Sea show $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios of 1.22 ± 0.11 (age corrected to 1/1/15), suggesting aged thermal fission product Cs discharged from Sellafield. The differences in $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios between Sellafield, Chernobyl, and global nuclear weapons testing fallout indicate that $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios can be utilized to discriminate between and track radiocesium transport from different nuclear production source terms, including major emission sources in Europe.

Keywords

Cs-135, Cs-137, Chernobyl, Sellafield, attribution

1. Introduction

Large quantities of anthropogenic radioactivity have been released to the environment as a result of industrial nuclear activities [Kayzar 2014; Markham 1978; Napier 2014; Farris 1996], medical isotope production [Bowyer 2013; Johnson 2015], accidents such as Chernobyl and Fukushima [UNSCEAR 2000; Buesseler 2011], and nuclear weapons testing [Krey 1973]. These releases have resulted in both high localized concentrations of anthropogenic radioactivity in the vicinity of some nuclear facilities/sites and very low levels of radioactivity dispersed both globally and regionally [UNSCEAR 2000, IAEA 2005, Krey 1973]. As radioactivity can be harmful to the biosphere, the long term effects of these radionuclide releases are currently being investigated.

Attribution of anthropogenic contamination to a given emission source term is critical to understanding how radionuclides are transported and distributed within the environment. While the elemental composition from a given nuclear source term is not always unique, the isotopic composition frequently varies as a function of the original nuclear source term's operating history and conditions; such variances introduce the possibility to discriminate between different contamination source terms based upon isotopic analyses [Zheng 2014; Taylor 2008; Snow 2015]. Furthermore, the unique isotopic "fingerprint" of different nuclear emission source terms can provide a tool for studying important environmental phenomena including atmospheric and ocean current patterns [IAEA 2005; Buesseler 2011], aeolian/pluvial/fluvial environmental transport [Snow 2015a], and soil erosion/sedimentation rates [Lee 1993].

^{137}Cs represents a major radionuclide of environmental concern as it is produced in high yield during nuclear fission (6.19% for thermal fission of ^{235}U [Baum 2002]), has a 30.07 yr half life, and is a strong β/γ emitter. As low levels of ^{137}Cs are ubiquitous throughout the environment as a result of nuclear weapons testing activities, discrimination between different environmental radiocesium source terms (e.g. Chernobyl, Sellafield, La Hague, global nuclear weapons testing fallout, etc.) requires analysis of other isotopes of cesium. Two other anthropogenic isotopes of Cs (e.g. ^{134}Cs and ^{135}Cs) are potentially useful for tracking radiocesium transport over the course of several years after release. While $^{134}\text{Cs}/^{137}\text{Cs}$ measurements are relatively easy to perform radiometrically, ^{134}Cs 's 2.065 yr half life limits $^{134}\text{Cs}/^{137}\text{Cs}$ ratios utility to ~10 years after a fission event [Snow 2015b; Zheng 2014]. On the other hand, ^{135}Cs has a much longer half life (2.3E6 yrs) and thus $^{135}\text{Cs}/^{137}\text{Cs}$ can provide a valuable tracer for much longer time periods; however, $^{135}\text{Cs}/^{137}\text{Cs}$ ratios are challenging to measure as ^{135}Cs 's long half life and low atomic abundance preclude simple radiometric detection. Current state-of-the-art $^{135}\text{Cs}/^{137}\text{Cs}$ ratio measurement techniques require extensive sample purification prior to analysis via ultra-high sensitivity mass spectrometry [Snow 2015b; Zheng 2014; Taylor 2008; Shibahara 2014; Russell 2015].

While many studies have reported ^{134}Cs and ^{137}Cs concentrations in soils and vegetation across Europe [Paatero 2007; Mietelski 2008; Duff 2008; Lehto 2008; Khrushchinskii 2014], to date only one study of soil samples within the Chernobyl exclusion zone has reported $^{135}\text{Cs}/^{137}\text{Cs}$ ratios [Taylor 2008]. As several different anthropogenic emission source terms in Europe exist, characterization of the general $^{135}\text{Cs}/^{137}\text{Cs}$ ratio distribution across Europe could

be of great value to future contamination source term analyses and environmental transport studies in this region.

In this work we demonstrate how $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios can be utilized to discriminate between and track environmental contamination from various nuclear production source terms. Environmental samples analyzed in this work represent a variety of different matrices (e.g. soil, clover, spinach, moss, seawater, and seaweed) taken from various distances and directions from major European anthropogenic emission source terms (including Chernobyl, Sellafield, La Hague, and global nuclear weapons testing fallout, see Figure 1). ^{137}Cs in environmental samples taken from the European mainland are anticipated to represent radiocesium derived from atmospheric deposition processes, while the Irish seawater samples are anticipated to primarily reflect discharges from reprocessing activities (including subsequent aqueous radiocesium resuspension/deposition processes [MacKenzie 1993; Tyler 2003; McCubbin 2002]). The results from this work demonstrate the utility for $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios to discriminate between different contamination source terms; furthermore, these results provide an enabling $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratio foundation supporting future European radiocesium environmental studies.

1.1. Expected Trends in ^{137}Cs Concentrations and $^{135}\text{Cs}/^{137}\text{Cs}$ Isotope Ratios From European Emission Source Terms

A brief discussion of anticipated variations in radiocesium concentrations and isotope ratios for major European emission source terms is given in this section in order to provide context for the results of this work; for in depth analyses of these factors the reader is referred

to the following references [UNSCEAR 2000; UNSCEAR 2008; Mackenzie 1993; McCubbin 2002; Snow 2015; Snyder 2012; Taylor 2008; Russell 2015].

Estimates of the total quantities of ^{137}Cs released/deposited from various source terms impacting Europe are shown in Figure 2. First, although global nuclear weapons testing fallout represents the single largest atmospheric ^{137}Cs emission source term to the environment, ^{137}Cs from this source was distributed globally (due to long residence times and mixing in the stratosphere prior to deposition, with larger quantities deposited to the northern hemisphere than the southern hemisphere [Bergan 2002; UNSCEAR 2000]) and thus the total quantity of ^{137}Cs within European surface soils/vegetation from this source term is anticipated to be relatively low (generally on the order of ~ 0.01 Bq/g, with variances at different locations based upon annual precipitation and other deposition factors [Monna 2009; Antovic 2012]). In contrast, atmospheric ^{137}Cs emissions from other European source terms such as Chernobyl, Sellafield, and La Hague were deposited regionally (due to negligible stratospheric penetration during release events), with exact deposition concentrations and patterns varying based upon meteorological conditions at the times of release [UNSCEAR 2000]. Thus, in general, surface soil/vegetation radiocesium concentrations in Europe are expected to reflect low levels of ^{137}Cs distributed ubiquitously from global nuclear weapons testing fallout, along with higher levels (~ 1600 Bq/g or greater in the Chernobyl exclusion zone [Barnett 2009]) of ^{137}Cs in the vicinity of major European emission source terms (including “hot spots” here fallout from the Chernobyl accident was preferentially deposited) [UNSCEAR 2000; UNSCEAR 2008].

Finally, $^{135}\text{Cs}/^{137}\text{Cs}$ ratios from each of the European source terms are hypothesized to be distinct from each other (based upon expected differences in the fission conditions and

average fission age from each source term [Snow 2015a; Robel 2015]); however, as only $^{135}\text{Cs}/^{137}\text{Cs}$ measurements of global nuclear weapons testing fallout and Chernobyl have been reported in the literature, this hypothesis still requires validation (and thus represents a primary objective of this work). Measurements of $^{135}\text{Cs}/^{137}\text{Cs}$ ratios from global nuclear weapons testing fallout by Lee et al., Chen et al., and Snyder et al. suggest an initial, fast fission $^{135}\text{Cs}/^{137}\text{Cs}$ production ratio of ~ 1.0 at the peak of nuclear weapons testing in 1964; this ratio corresponds to an age corrected ratio of ~ 3.2 as of 1/1/2015 [Lee 1993; Chen 2008; Snyder 2012]. On the other hand, measurements of soils within the Chernobyl exclusion zone by Taylor et al. show a $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratio of 0.30 ± 0.02 decay corrected to April 26, 1986, the date of the accident (0.58 ± 0.04 decay corrected to 1/1/2015 [Taylor 2008]). While to our knowledge $^{135}\text{Cs}/^{137}\text{Cs}$ ratio measurements have not been reported to date for material from Sellafield or La Hague, the majority of material reprocessed by these facilities is believed to have been derived in thermal to epithermal fission events (as fuels reprocessed at Sellafield and La Hague came primarily from moderated reactor types including light water reactors and various Magnox and other graphite moderated gas reactor designs [Mogg 1987; Merry 1995; Nuclear Quality Knowledge 2013; Schneider 2008]). Thus, emissions from the Sellafield and La Hague reprocessing facilities are anticipated to broadly reflect aged thermal fission product signatures (estimated integrated ratio of ~ 0.2 to 0.5 at the time of fission, with exact $^{135}\text{Cs}/^{137}\text{Cs}$ ratio in 1/1/2015 varying according to the exact fission date). For Sellafield, such a ratio would be predicted to range anywhere from ~ 0.33 up to ~ 2.23 (where an initial thermal fission product ratio of 0.2 - 0.5 is predicted and a fission date uncertainty spanning from 1950 when the first nuclear reactor became operational at Sellafield to 1993 when the seawater samples

was collected), with an average ratio range in 1/1/2015 for Sellafield of 0.51-1.29 predicted for peak discharges in 1975.

2. Method

2.1. Cesium Purification

Environmental samples were collected by the International Atomic Energy Agency (IAEA) from 1986 to 2006; details regarding the sample collection, homogenization, and treatment are reported elsewhere [Strachnov 1996; Strachnov 1991; IAEA 2009a; IAEA 2009b; IAEA 2009c]. Upon receipt at INL, samples underwent different initial treatments based upon their specific matrix. Soil samples were dry ashed at 550 °C for 20 hours, following which 0.6 g of the dry ashed material were added to individual CEM MARS 6® Microwave Digestion vessels; for most vegetation samples, 0.3 g of sample were added to individual microwave vessels without prior treatment. Dry ashing soil samples was required to completely digest these matrices and was not observed to result in a measurable decrease in the ^{137}Cs activities in the samples analyzed in this work (within $\sim 5\%$ 2σ ^{137}Cs activity measurement uncertainties). Furthermore (and more importantly), $^{135}\text{Cs}/^{137}\text{Cs}$ isotopic analyses of vegetation samples that had been processed through the dry ashing step showed statistically identical $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios to samples that had not been dry ashed (within the $\sim 2\text{-}4\%$ 2σ measurement precision limit we are currently able to attain).

After sample addition to individual microwave vessels, 4 mL of 16 M HNO_3 and 4 mL 29 M HF were added to each microwave vessel, following which vessels were sealed and microwaved using the procedure of Snow et al. [Snow 2015b] Upon cooling, an additional 30

mL of 44 g/L boric acid was added to each vessel, following which vessels were resealed and remicrowaved. In the case of seawater samples, microwave digestion was not performed; 100 mL of 16 M HNO₃ was added directly to 1 L of seawater and samples were filtered through a 0.45 µm filter. After microwave digestion (soils and vegetation) or acid addition and filtration (seawater), samples underwent purification via two ammonium molybdophosphate polyacrylonitrile columns, two anion exchange separations (AG 1x8 resin, OH⁻ form), and a final cation exchange column (AG50W resin) prior to analysis via thermal ionization mass spectrometry (TIMS), with specific details regarding the chemical purification method reported elsewhere in the literature [Snow 2015b].

2.2. Method Validation

No ¹³⁵Cs/¹³⁷Cs standard is currently commercially available; however, several groups have reported values for the IAEA-375 reference material. Comparison between values obtained by our lab for this material and those reported in the literature is given in Table 1; data in this study is in quantitative agreement with values reported by the two other reporting laboratories. Furthermore, quantitative agreement between our group and that of Taylor et al. enables direct comparison of data from this study to that reported by Taylor et al. for samples taken from within the Chernobyl Exclusion Zone [Taylor 2008].

3. Results and Discussion

3.1. Atmospheric ¹³⁵Cs/¹³⁷Cs Deposition Across Eastern and Central Europe

Table 2 shows the experimentally measured $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratio values from European samples. With the exception of the Irish Seawater samples and one moss-soil sample from Hungary, all samples show statistically similar $^{135}\text{Cs}/^{137}\text{Cs}$ values ranging from 0.50 to 0.64. These values are found to be in quantitative agreement with values reported by Taylor et al. from soil samples within the Chernobyl Exclusion Zone ($^{135}\text{Cs}/^{137}\text{Cs}$ of 0.58 ± 0.04 as of 1/1/2015), strongly suggesting that the general $^{135}\text{Cs}/^{137}\text{Cs}$ isotopic composition at the sampling locations in this work represents atmospheric deposition from the Chernobyl nuclear power plant (and that other nuclear source terms such as global nuclear weapons testing fallout represent insignificant components to the radiocesium at the specific sample locations in this work).

One interesting observation is that the $^{135}\text{Cs}/^{137}\text{Cs}$ isotopic composition of radiocesium from Chernobyl appears to be homogenous irrespective of distance or direction from the Chernobyl nuclear power plant. This observation is of particular interest because the $^{135}\text{Cs}/^{137}\text{Cs}$ isotopic composition within moderated nuclear reactors is not expected to be uniform throughout the reactor core; rather, in light water reactors $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios correlate with neutron flux profiles, with $^{135}\text{Cs}/^{137}\text{Cs}$ values of ~ 1.0 at the ends of the fuel rods (where neutron fluxes are lowest) and depleted $^{135}\text{Cs}/^{137}\text{Cs}$ values toward the center of the fuel rods (where fluxes are highest) as described in detail by Snow et al. and Robel et al. [Snow 2015c, Robel 2015]. Thus, depending on the circumstances surrounding the release event, one might expect to see variances in the $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratio emitted during a reactor accident as a function of time based upon the fuel position, extent of core damage, and meteorological conditions at the time of release [Snow 2015c]. Interestingly, while meteorological conditions

varied dramatically over the course of hours to weeks following the Chernobyl accident [UNSCEAR 2000], variation in the environmental $^{135}\text{Cs}/^{137}\text{Cs}$ ratio as a function of distance and direction from Chernobyl is not observed. One explanation that accounts for this is that accident conditions at Chernobyl resulted in sufficiently high temperatures and widespread damage to result in near complete volatilization and homogenization of gaseous radiocesium within the bulk of the reactor core; such a hypothesis is consistent with the known accident scenario (where the central portion of fuel within the reactor core is believed to have been nearly instantaneously vaporized during the rapid power excursion, along with cladding failure and melting of the remainder of the core) [UNSCEAR 2000].

While most European surface soil and vegetation samples in this study appear to have very consistent $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios, a single sample from Hungary showed a statistically higher $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratio. It is possible that this sample contained higher quantities of global nuclear weapons testing fallout ($^{135}\text{Cs}/^{137}\text{Cs}$ ratio of ~ 3.2 as of 1/1/2015) or an anomalous “hot particle” from a peripheral location within the Chernobyl core, however such explanations are deemed unlikely (though perhaps still possible) as ^{137}Cs activity analyses of replicate samples from the same sample batch suggest that the ^{137}Cs concentration within this batch was homogenous (see Figure 3). Thus, at this date the source of this higher ratio in this particular sample is unknown.

3.2. Irish Seawater

$^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios from the Irish Seawater samples are shown in Table 2. $^{135}\text{Cs}/^{137}\text{Cs}$ ratios from all three replicates are statistically similar to each other, with the highest

precision value measured at 1.21 ± 0.04 (decay corrected to 1/1/2015). Importantly, $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratio measurements from the Irish Seawater are statistically different from both Chernobyl (0.58 ± 0.04) and global nuclear weapons testing fallout (~ 3.2 [Snyder 2012]). Due to the close proximity to the Sellafield reprocessing facility, where very large quantities of radiocesium are known to have been released to the Irish Sea [UNSCEAR 2008; McCubbin 2002; MacKenzie 1993], the measured $^{135}\text{Cs}/^{137}\text{Cs}$ within the Irish Seawater samples are determined to most likely be derived from radiocesium releases from the Sellafield reprocessing facility. Importantly, the difference in $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios for Sellafield, Chernobyl, and global nuclear weapons testing demonstrates that $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios can be utilized to discriminate between different European nuclear emission source terms, and thus can provide a valuable tracer for tracking radiocesium transport and mixing between each of these source terms as a function of time (particularly in major regions of mixing such as the western Baltic Sea).

Radiocesium chemistry within the Irish Sea has been well documented; upon release $\sim 90\%$ of the radiocesium remains in the aqueous phase and is transported out of the Irish Sea within 1 year, while the remaining $\sim 10\%$ of the radiocesium is scavenged by particulates (predominantly fine sediments and clay particles) and deposited to the sea floor [MacKenzie 1993]. Interestingly, while large scale discharges from Sellafield to the Irish Sea ceased in 1985, continuous low level ^{137}Cs concentrations in seawater continue to be observed; these residual low levels of ^{137}Cs have largely been attributed to resuspension of previously deposited legacy radiocesium from Sellafield [MacKenzie 1993; Tyler 2003; McCubbin 2002]. As the Sellafield reprocessing facility primarily reprocessed fuel from moderated reactors (e.g. various light

water, Magnox, and graphite moderated gas reactor designs [Mogg 1987; Merry 1995; Nuclear Quality Knowledge 2013; Schneider 2008]), legacy radiocesium from Sellafield is anticipated to have an initial $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratio representative of thermal fission product cesium (e.g. < 1.0, generally in the 0.2 to 0.5 range at the time of production). Since the Irish Sea samples in this work were collected in September 1993, radiocesium within these samples must have been produced in 1993 or earlier; this results in a calculated initial $^{135}\text{Cs}/^{137}\text{Cs}$ ratio of 0.74 ± 0.02 (if produced in 1993) or lower (if produced prior to 1993). Interestingly, when $^{135}\text{Cs}/^{137}\text{Cs}$ ratios are decay corrected to the peak of seawater discharges (1975) [UNSCEAR 2000], a ratio of 0.48 ± 0.02 is calculated; this ratio is within the range roughly predicted for pure thermal fission product cesium produced in 1975 (estimated $^{135}\text{Cs}/^{137}\text{Cs}$ ratio range of 0.2 to 0.5 decay corrected to 1/1/1975, see section 1.1.).

4. Conclusions

Surface soil and vegetation samples from central and eastern Europe show consistent $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratio values ranging from 0.50 to 0.64; these values are in statistical agreement with previous measurements from soils within the Chernobyl exclusion zone (0.58 ± 0.04 [Taylor]), suggesting that radiocesium at the locations in this work is derived primarily from Chernobyl. In the case of the Irish Seawater samples, a $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratio of ~ 1.2 is observed; decay correction of this ratio to the time of sampling indicates that radiocesium within the Irish Sea is derived from thermal fission event(s) and is most likely derived from reprocessing releases from the Sellafield reprocessing plant. Importantly, $^{135}\text{Cs}/^{137}\text{Cs}$ ratios measured from Chernobyl and Sellafield are observed to be distinct from each other and from

ratios previously measured for global nuclear weapons testing fallout [Snyder 2012], indicating that $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios can potentially provide a valuable tool for tracking radiocesium transport and mixing between each of these source terms as a function of time.

While this work provides a preliminary foundation for future European radiocesium environmental transport and mixing studies, it is worth mentioning several potential areas of future research. First, while $^{135}\text{Cs}/^{137}\text{Cs}$ isotopic emissions from Chernobyl appear to be very homogenous, the $^{135}\text{Cs}/^{137}\text{Cs}$ isotopic homogeneity of Sellafield releases to the Irish Sea is not currently known. As discharges from various fuel batches from Sellafield occurred intermittently to the Irish Sea, it is possible that localized variations in $^{135}\text{Cs}/^{137}\text{Cs}$ at the seafloor exist. Thus, more widespread analyses of both Irish Seawater and sediment samples could be of benefit in more precisely evaluating the temporal and geographic homogeneity of Sellafield derived radiocesium and its associated $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratio as a mixing model end member. Interestingly, these intermittent discharges from Sellafield as a function of time could possibly enable reconstruction of Sellafield's release history based upon $^{135}\text{Cs}/^{137}\text{Cs}$ analyses of Irish Sea sediment cores as a function of depth, though this hypothesis would need to be evaluated. Finally, while releases from La Hague were much lower than other radiocesium source terms in Europe (Figure 2), analyses of samples in the vicinity of this facility could also be of value in more precisely defining the $^{135}\text{Cs}/^{137}\text{Cs}$ isotopic composition of the La Hague end member (though La Hague's impact on European radiocesium as a whole is anticipated to be much smaller than both Chernobyl and Sellafield).

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Figure 1. Locations of the samples analyzed in this work compared with major European ^{137}Cs emission source terms (note: IAEA-156 clover samples were taken in a small, unspecified geographical region of Austria [Strachnov 1991], thus the point in this map for IAEA-156 represents an approximate location).

Figure 2. Release histories from various a) atmospheric and b) seawater discharge source terms across Europe. Data for nuclear weapons fallout represents ^{137}Cs deposition estimates, while data for Sellafield, La Hague, and Chernobyl represent emission estimates. Discharges from Sellafield and La Hague prior to 1970 are known to have occurred, however the total quantities released have not been reported. Data adapted from UNSCEAR 2000 and UNSCEAR 2008 reports [UNSCEAR 2000; UNSCEAR 2008].

Figure 3. Gamma spectral analyses of separate 3g fractions of IAEA-447 (Hungarian soil-moss). Samples 4-1 through 5-2 represent individual 5 g subsamples of IAEA-447. Error bars represent 2 σ error.

Table 1. Method Validations using IAEA-375. $^{135}\text{Cs}/^{137}\text{Cs}$ ratios are decay corrected to 4/26/1986. Sample sizes in this study ranged from 1.5 g to 7.2 g initial sample. Error bars represent 2σ error.

IAEA-375	$^{135}\text{Cs}/^{137}\text{Cs}^*$
This Work (n = 7)	0.30 ± 0.01
Taylor et al. (n=3)	0.30 ± 0.03
Zheng et al. (n=3)	0.31 ± 0.01

Table 2. ^{137}Cs concentrations and $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratios from samples collected across Europe, compared with ratios measured from global nuclear weapons testing fallout and predicted ratios for thermal fission derived radiocesium for a given range of production date. Errors represent 2σ .

Sampling Location	Sampling Date	Matrix	^{137}Cs (Bq/g)	Sample #	$^{135}\text{Cs}/^{137}\text{Cs}$ (atoms)
Novozybkov, Russia (IAEA-375)	July 1990	Soil	3.11 ± 0.09	#1	0.59 ± 0.04
				#2	0.55 ± 0.04
				#3	0.58 ± 0.02
				#4	0.62 ± 0.03
				#5	0.58 ± 0.02
Kiev, Ukraine (IAEA-330)	Summer 1990	Spinach	1.05 ± 0.06	#1	0.56 ± 0.02
				#2	0.57 ± 0.03
Austria (IAEA-156)	Summer 1986	Clover	0.14 ± 0.01	#1	0.58 ± 0.04
				#2	0.58 ± 0.02
				#3	0.57 ± 0.02
Northwest Hungary (IAEA-447)	Summer 1987	Moss-Soil	0.38 ± 0.02	#1	0.58 ± 0.08
				#2	1.78 ± 0.12
Irish Sea (IAEA-443)	Sept. 1993	Seawater	0.00030 ± 0.00002	#1	1.18 ± 0.12
				#2	1.21 ± 0.04
				#3	1.27 ± 0.31
Chernobyl [Taylor 2008]	1989	Soil	14-46	-	0.58 ± 0.04
Global Nuclear Weapons Testing Fallout [Snyder 2012]	1964	Soil	-	-	~ 3.2
Thermal Fission between 1952 and 1993 (initial $^{135}\text{Cs}/^{137}\text{Cs}$ ratio 0.2-0.5)	-	-	-	-	~ 0.33 to 2.23
Thermal Fission in 1975 (peak of Sellafield discharges)	-	-	-	-	~ 0.51 to 1.29

* All ^{137}Cs activity and $^{135}\text{Cs}/^{137}\text{Cs}$ isotope ratio data are decay corrected to 1/1/2015.

** The column "Sample #" refers to different replicate digestions and subsequent analyses of the same original parent material

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Graphical Abstract