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Prediction of I-131 Influence in the Aegean Region by Chernobyl Accident Using the Ratio of I-129/I-127 in the Lake Sediments

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ABSTRACT: After the Chernobyl accident, as in many countries, the short half-life I-131 radioactivity could not be measured in Turkey-Aegean Region. By using Cs-137 radioactivity, which is easy to monitor, I-131 radioactivity was tried to be predicted, but it was thought that the results obtained by these two radioisotopes because of their different chemical properties would not be correct. In this study I-129 was studied to predict the retrospective I-131 radioactivity. I-129, another iodine radioisotope, was thought to be more appropriate as a predictive agent because of the same chemical properties as I-131 and also its long half-life. I-129 was measured in the Bafa Lake sediments by using microwave digestion, extraction and AMS methods and retrospective I-131 activity was predicted. As a result, it was found that I-131 radioactivity values in Bafa Lake habitat is between 9.78x10⁻³ Bqkg⁻¹ and 1.02x10⁻² Bqkg⁻¹ and the accuracy of the method used has been proved.

Keywords: Chernobyl accident, Bafa Lake, I-129, I-131, AMS, lake sediment

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INTRODUCTION

As it is known, a big explosion occured in the 4th reactor of the Chernobyl Nuclear Power Plant in Ukraine-Chernobyl region on April 26, 1986 (TAEKa, 2007). The radioactive particles that started to leak into the atmosphere as a result of the explosion were first affected the countries of Ukraine, Belarus and the Russian Federation, and then Northern European countries such as Finland, Sweden and Norway. According to Turkey State Meteorology Affairs Directorate report of wind and regional precipitation for 1-9 May 1986, it is believed that Turkey also seriously affected by the radioactive contamination (TAEKb, 2007). However, as in many countries, I-131 radioactivity could not be measured in many parts of our country, especially in the Aegean Region due to its short (8.02 days) half-life (TAEKc, 2007).

After a nuclear accident, radioactive fallout is transported to the lake environment by dry/wet deposition, streams and erosion of contaminated soils. Lakes are suitable for retrospective isotope measurements due to the being a sealed system aganist readionuclides. Thus, in this study, the amount of I-129 was measured in the Bafa Lake in the Aegean Region in order to foresee the I-131 activity influenced to the Aegean Region in the accident. In this study it was also aimed to discuss the accuracy of the method and contribute to the literature.

I-131 is important for environmental contamination caused by radioactive waste due to its volatility, short half-life and abundance in fission products. In addition, I-131 is an important radioactive isotope for human health, as it can accumulate in thyroids and cause an increase in thyroid cancer.

The release of the I-131 radionuclide in Chernobyl took place over a period of 10 days at variable speeds as indicated in Figure 1 (TAEKa, 2007).





Some amount of radionuclides released into the atmosphere in the Chernobyl accident are stated as 6 kg for I-129 (Aldahan et al., 2007) and 1200-1760 PBq for I-131 (TAEKa, 2007; UNSCEAR, 2008).

Then, I-131 activity was tried to be predicted by making use of Cs-137 radioactivity, which is easy to monitor due to its long half life (30.17 years). However, the accuracy of the obtained I-131 activity results by this method is controversial due to the different chemical behaviour of the two radioisotopes.

Soil, water and plant samples of different countries contaminated after the accident were studied and the I-131 amount was tried to be found by using the atom ratios of I-129/Cs-137, I-131/Cs-137 (Dubina et al., 1990; Mironov et al., 2002; Hou et al., 2003; Michel et al., 2005).

I-129 concentrations were examined in 42 soil profile samples in 1995 and 1997, respectively, in contaminated soils of Russia and Ukraine. It is found that I-129 activities varied in the range of 0.06-

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24.5 mBqkg⁻¹ depending on the depth factor in soil profiles. In just two examples, a very high activity of I-129 such as 52 mBqkg⁻¹ in the Russia-Gomel region and 111 mBqkg⁻¹ at 800 meters from Chernobyl were detected. In similar studies, I-129/I-131 atom ratio was found to be 15.2±4.7 in 24 soil samples (Mironov et al., 2002) and I-129/I-131 atom ratio was found to be 2±10 in animal thyroids (Van Middlesworth et al., 1997). With the help of these data, the detection of the migration routes of iodine isotopes and the feasibility of calculating the historical dosimeter of I-131 has been proved (Fehn et al., 2002). Then, I-129 and I-131 measurements were made in soil, seawater and pelagic sediment, animal thyroid, lake water/sediment, air and plant samples by many different researchers in different parts of the world. In the studies, using the I-129/I-131, I-129/I-127 atom ratios, contamination of the regions caused by nuclear accidents and migration routes of iodine were investigated (Edwards vd 1962; Fabryka-Martin et al., 1985; Arntsing et al., 1991; BstMLU, 1998; Mironov et al., 2002; Ernst et al., 2003; Hou et al., 2003; Michel et al., 2005; Matsuzaki et al., 2007; Englund et al., 2008; Liu et al., 2008; Muramatsu et al., 2008; Otosaka et al., 2008; Hou et al., 2009; Englund et al., 2010; Hou et al., 2010; Zhang et al., 2011; METX, 2011; Daraoui et al., 2012; Miyake et al., 2012; Miyake et al., 2015; Muramatsu et al., 2015; Ezerinskins et al., 2016; Fan et al., 2016; Matsunaka et al., 2016; Yang et al., 2017; Zhang et al., 2018).

In their studies, Daraoui et.al. reported that Cs-137 and I-129 showed different distributions in sediment profile, while Michel et.al. reported that I-129 and Cs-137 mobility was different in the soil (Daraoui et.al. 2012; Michel et.al. 2015). Miyake et.al. reported that I-129 and I-131 will exhibit similar chemical behavior in the environment and that I-131 activity can be predicted by performing I-129 measurement (Miyake et al., 2015). In the study reported by Fujiwara (Fujiwara, 2016) the temporal variations of I-131 deposited on the ground and accumulated in cropland soil during the early stages of the Fukushima nuclear accident were monitored. I-129 concentrations in atmospheric deposits and soil were measured to examine the feasibility of retrospectively reconstructing I-131 levels from the levels of accident-derived I-129 levels and the I-131 concentrations measured in the topsoil were reported as very consistent with the I-131 concentrations reconstructed from the I-129 concentrations in the soil.

In this study, it is thought that the use of Cs-137 measurements to predict retrospective I-131 activity will not be correct, due to different chemical behaviors of cesium and iodine in soil, water and sediment environments. Instead, I-131 amounts were predicted by using I-129 measurements that has the same chemical properties and behaviors with I-131. Thus accuracy of the method was questioned and the I-129 and I-131 activities were studied for Turkey-Egean Region fort he first time.

MATERIALS AND METHODS

Sampling Area

Bafa Lake is a natural embankment lake with a surface area of 65 km² located in the coordinates of 37 31' North, 27 27' East (Googlemaps, 2019; Balık and Ustaoğlu, 1989). The depth of the lake reaches 21 m in some places and the percentage of sludge in its sediments varies between 6 and 97%. The sediments in the center of the lake are storage centers with sludge amounts exceeding 90% (Yılgör, 2012).

The Bafa Lake sedimentation rate varies between 0.089-7.587 cm and the sedimentation rate is on average 0.36 cm per year (Yılgör, 2012). In our study, sediment samples were taken on April 11, 2017 and 31 years have passed since the accident. In this case, approximately 11.408 cm sediment has accumulated in Lake Bafa after the accident.

Sampling and Pretreatment

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As shown in Figure 2, samples were taken from 5 different points at a depth of 5-10 m with a UWT Core sediment device as a 60 cm sediment profile sample. Sample coordinates are given in Table 1. The samples taken were kept at 4^{0} C.



Figure 2. Lake Bafa sampling points

Sediment samples were divided into slices in 2 cm layers and dried at 103.5 ^oC for 24 hours. The weight of each sample was sensitively measured. Dry weights of the samples were found to be 16 g on average. Samples were then ground in a ball mill and sieved to a particle size of <77 microns.

Table 1. Lake Bafa sampling coordinates

Sample Codes					
	N1	N2	N3	N4	N5
Coordinates	37 ⁰ 31'5.8" N	37 ⁰ 29'54.6" N	37 ⁰ 29'46.1" N	37 ⁰ 29'44.1" N	37 ⁰ 29'43.8" N
(North/East)	27 ⁰ 27'52.4" E	27 ⁰ 26'53.8" E	27 ⁰ 27'39.5" E	27 ⁰ 28'23.0" E	27 ⁰ 29'9.5" E

Taking Iodine in Sediment into Solution Medium

Microwave digestion method was used to take iodine into solution medium. 0.5 g sediment sample was taken into the PTFE-TFM container and 2 mg iodine was added to facilitate the migration of the isotopes in the sample to the final step. As the solvent, 10 mL of 60% HNO₃ was added then waited until CO₂ output due to the interaction of organic matter and acid was end. The samples were then subjected to digestion in the microwave (Gomez-Guzman, 2011). The ramp time for digestion was determined as 10 min, waiting time as 20 min and target temperature as 200°C. After digestion, the samples were filtered, the solution volume was completed to 50 ml and extraction was started.

Extraction was done by repeating 3 times with 12 ml of chloroform. 0.1 M 20 ml of sodium bisulphite solution was added to the chloroform phase. Then 0.1 M 20 mL of bariumnitrate solution was added to precipitate sulfate and sulfite ions in solution medium. Centrifugation for 20 minutes at 4000 rpm allowed the sulfate and sulfite ions in the solution to settle as $Ba(SO_4)_2$. 0.1 M 600 µL silver nitrate solution was added to precipitate the iodine, which is oxidized in the separated solution. The color of the solution, which was colorless at this stage, turned cloudy yellow, indicating that the iodine precipitate was dried at 60°C for 24 hours. In the last step, as the excess silver nitrate added collapsed as silver oxide,

blackish gray color was observed in the precipitate. Then atomic mass spectrometry measurements of the dried precipitate were made.

Atomic Mass Spectrometry (AMS) Measurements

AMS measurements were performed with 1MV HVEE AMS at the Romanian-Bucharest University Horia Hulubei National Institute of Physics and Nuclear Engineering. The properties of the used AMS system are given in Table 2.

Table 2. Properties of the AMS system used in the study

Size	4.2x6.2 m
Ion source	100 μ A max.–2 seperated injector 120 ⁰
Tension before acceleration	35 kV
Injector magnet	3 kV, 100 Hz
Terminal voltage and charging current	1 MV, 2 mA
Analysis magnet	90 ⁰ , 63 MeV amu, max current 300 A
Electric global analyzer	$(ESA) 120^{0}, \pm 60 \text{ kV}$

RESULTS AND DISCUSSION

I-129 Activity Calculations

In the study, the data in terms of particles g⁻¹ obtained with AMS were expressed in Bq for each layer sample, in order to make the results meaningful. For this purpose, I-129 activities were calculated by using Equation 1.

 $A = N \times \lambda$

In Equation 1., A represents activity in Bq, N represents number of radioactive particles and λ represents decay constant in (s⁻¹). λ value of I-129 was calculated by using Equation 2. where $t_{\frac{1}{2}}$ is half life of I-129 that equals to 1.57×10^7 years.

$$\lambda=0,693/t_{\frac{1}{2}}$$

Sample codes, depths, AMS results (particles g⁻¹) and calculated I-129 activities (Bqg⁻¹, Bqkg⁻¹) are given in Table 3.

Table 3. Sample codes, depths, AMS results and calculated I-129 activities (nd: undetectable through the used analysis method

Sample code	Depth (cm)	Particle number (particles g ⁻¹)	Activity (Bqkg ⁻¹)	Sample code	Depth (cm)	Particle number (particles g ⁻ ¹)	Activity (Bqkg ⁻¹)
	(10-12)	1.71×10^{8}	2.39x10 ⁻⁴	_	(10-12)	Nd	Nd
	(12-14)	Nd	Nd	_	(12-14)	2.03×10^{8}	2.84x10 ⁻⁴
N1	(14-16)	9.67x10 ⁷	1.35x10 ⁻⁴	N2	(14-16)	Nd	Nd
	(16-18)	5.76x10 ⁷	8.06x10 ⁻⁵	_	(16-18)	Nd	Nd
	(18-20)	Nd	Nd		(18-20)	Nd	Nd
	(10-12)	1.51x10 ⁸	2.11x10 ⁻⁴	_	(10-12)	2.29×10^8	3.21x10 ⁻⁴
	(12-14)	Nd	Nd	_	(12-14)	Nd	Nd
N3	(14-16)	9.18x10 ⁷	1.29x10 ⁻⁴	N4	(14-16)	Nd	Nd
	(16-18)	Nd	Nd	_	(16-18)	Nd	Nd
	(18-20)	Nd	Nd		(18-20)	Nd	Nd
	(10-12)	Nd	Nd	_			
	(12-14)	Nd	Nd	_			
N5	(14-16)	5.00×10^7	7.00x10 ⁻⁵	_			
	(16-18)	Nd	Nd	_			
	(18-20)	Nd	Nd				

(1)

(2)

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In order to evaluate the results more easily, the obtained data were grouped. Also, since the average sediment accumulation rate of Bafa Lake, which is the sample area in the study, is approximately 0.387 cmyear⁻¹ (Y1lgör, 2012), the age determination of the sediment layers was made based on the layer depth. The results obtained are given in Table 4.

Sample Code	Depth:18-20 cm Age: 47-52 years Date: 1965-1970	Depth:16-18 cm Age: 41-47 years Date: 1970-1976	Depth:14-16 cm Age: 36-41 years Date:1976-1981	Depth:12-14 cm Age: 31-36 years Date:1981-1986	Depth:10-12 cm Age: 26-31 years Date:1986-1991
N.5	Nd	Nd	7.00x10 ⁻⁸	Nd	Nd
N.4	Nd	Nd	Nd	Nd	3.21x10 ⁻⁷
N.3	Nd	Nd	1.29x10 ⁻⁷	Nd	2.11x10 ⁻⁷
N.2	Nd	Nd	Nd	2.84x10 ⁻⁷	Nd
N.1	Nd	8.06x10 ⁻⁸	1.35x10 ⁻⁷	Nd	2.39x10 ⁻⁷

Table 4. Distribution of I-129 activi	ty concentrations (Bqg	⁻¹) in	depth	profile
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Figure 3 was also obtained by converting the data given in Table 4 into a graphic.



Figure 3. Distrubition of I-129 concentration in depth profiles

When Figure 3 is interpreted, it was estimated that 1986, when Chernobyl accident occurred, corresponded to approximately 15 cm sediment depth. It was seen that this result is in accordance with the sediment layer dating by Yılgör (Yılgör, 2012). The reason for the deviations that can be ignored is the variability of the sediment accumulation rate in the lake and the difference of the iodine-holding capacity of the sediment.

As can be seen in Figure 3, it was observed that the I-129 accumulation was started above 15 cm depth that is corresponding 1987 (one year after accident) and reached a maximum in 1988-1990 and then I-129 activity decreased gradually.

The distributions of I-129/I-127 atom ratios in depth profiles, obtained from the experimental study results, are given in Table 5 and Figure 4.

Using the data in Figure 4 and Table 5, it is thought that the increase in the accumulation of I-129 in the post-1986 period was due to the Chernobyl accident induced fallout. The low concentration of I-129 concentration in the pre-1986 period is estimated to be due to nuclear fuel reprocessing facilities and nuclear weapons trials.

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Sample Code	Depth:18-20 cm Age: 47-52 years Date: 1965-1970	Depth:16-18 cm Age: 41-47 years Date: 1970-1976	Depth:14-16 cm Age: 36-41 years Date:1976-1981	Depth:12-14 cm Age: 31-36 years Date:1981-1986	Depth:10-12 cm Age: 26-31 years Date:1986-1991
N.5	Nd	Nd	3.10 x10 ⁻¹²	Nd	Nd
N.4	Nd	Nd	Nd	Nd	9.65 x10 ⁻¹²
N.3	Nd	Nd	4.40 x10 ⁻¹²	Nd	9.40 x10 ⁻¹²
N.2	Nd	Nd	Nd	1.26 x10 ⁻¹¹	Nd
N.1	Nd	3.80x10 ⁻¹²	5.10 x10 ⁻¹²	Nd	$1.00 \text{ x} 10^{-11}$





Figure 4. Distrubition of I-129/I-127 atom ratios in depth profile

The obtained results were compared with the literature values to question the accuracy of the method used in the study and the results are given in Table 6.

Doforonco	Sampling region,	Mothod and aquinmont	I-129 activity (Bqkg ⁻¹)		
Kelefence	Sample type	Method and equipment	Minimum	Maximum	
This study	Lake Bafa, Sediment	Microwave digestion, AMS	8.06x10 ⁻⁵	1.29x10 ⁻⁴	
Michel et al., 2005	Russia/Ukraine, Soil	RNAA, AMS	6.00x10 ⁻⁵	2.45x10 ⁻²	
Muramatsu et al., 2008	Japan, Soil	Pyrohydrolysis, AMS	1.40x10 ⁻⁵	4.50x10 ⁻³	
Daraoui et al., 2012	Germany, Soil	Standard method, AMS	6.20x10 ⁻⁴	5.40x10 ⁻³	
Daraoui et al., 2012	Chile, Soil	Standard method, AMS	1.50x10 ⁻⁴	1.62x10 ⁻³	
Ezerinskis et al., 2016	Lithuania, Forest soil	Pyrohydrolysis, AMS	2.81x10 ⁻⁴	1.43x10 ⁻³	
Ezerinskis et al., 2016	Lithuania, Meadow soil	Pyrohydrolysis, AMS	6.58x10 ⁻⁴	4.78x10 ⁻³	
Zhang et al., 2018	Philippines, Soil	Pyrohydrolysis, AMS	7.00x10 ⁻⁸	3.00x10 ⁻⁵	

 Table 6. Comparasion of results with literature values

As can be seen from Table 6, the results obtained in this study are in agreement with the literature data.

Prediction of I-131 Activity by Using I-129 Activity

It was suggested by Aldahan et al. (Aldahan et al., 2007) that 6 kg of I-129, whose activity was 4.10x10¹⁰ Bq (Equation 1.), was released into the atmosphere during the Chernobyl accident. It was reported by UNSCEAR that 1200-1760 PBq I-131 was released into the atmosphere during the accident (UNSCEAR, 2008). These values were used when predicting I-131 activity.

Table 7 contains the atom ratios of I-129/I-131 quantities of different studies given in the literature.

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Table 7. Atom ratios of 1-129/1-131 amounts in the interature			
Reference	I-129/I-131 (g/g)		
Mironov et.al. 2002	15.2		
VanMiddlesworth et.al. 1997	27.0		
Muramatsu et.al. 2015	19.5		

The amounts of I-129 by gram were calculated from the I-129 particle values in Table 3. The amount of I-131 by gram were calculated by using I-129/I-131 atom ratios given in Table 7. In addition, the activity (Bqkg⁻¹) values calculated using Equation 1. are also shown in Table 8.

Samle Code	Depth	I-131 (g)			1-131 (Bqkg ⁻¹)		
		Mironov et.al. 2002	VanMiddleswort h et.al. 1997	Muramatsu et.al. 2015	Mironov et.al. 2002	VanMiddlesworth et.al. 1997	Muramatsu et.al. 2015
N1	(10-12 cm)	2.41x10 ⁻¹⁵	1.36x10 ⁻¹⁵	1.88x10 ⁻¹⁵	1.11x10 ⁻²	6.23x10 ⁻³	8.63 x10 ⁻³
	(12-14 cm)	Nd	Nd	Nd	Nd	Nd	Nd
	(14-16 cm)	1.36x10 ⁻¹⁵	7.67x10 ⁻¹⁶	1.06x10 ⁻¹⁵	6.26x10 ⁻³	3.52x10 ⁻³	4.88x10 ⁻³
	(16-18 cm)	8.12x10 ⁻¹⁶	4.57x10 ⁻¹⁶	6.33x10 ⁻¹⁶	3.73x10 ⁻³	2.10x10 ⁻³	2.91x10 ⁻³
	(18-20 cm)	Nd	Nd	Nd	Nd	Nd	Nd
N2	(10-12 cm)	Nd	Nd	Nd	Nd	Nd	Nd
	(12-14 cm)	2.86x10 ⁻¹⁵	1.61x10 ⁻¹⁵	2.23x10 ⁻¹⁵	1.31x10 ⁻²	7.40 x10 ⁻³	1.02x10 ⁻²
	(14-16 cm)	Nd	Nd	Nd	Nd	Nd	Nd
	(16-18 cm)	Nd	Nd	Nd	Nd	Nd	Nd
	(18-20 cm)	Nd	Nd	Nd	Nd	Nd	Nd
N3	(10-12 cm)	2.13x10 ⁻¹⁵	1.20x10 ⁻¹⁵	1.66x10 ⁻¹⁵	9.78x10 ⁻³	5.50x10 ⁻³	7.62x10 ⁻³
	(12-14 cm)	Nd	Nd	Nd	Nd	Nd	Nd
	(14-16 cm)	1.29x10 ⁻¹⁵	7.29x10 ⁻¹⁶	1.01x10 ⁻¹⁵	5.94x10 ⁻³	3.35x10 ⁻³	4.63x10 ⁻³
	(16-18 cm)	Nd	Nd	Nd	Nd	Nd	Nd
	(18-20 cm)	Nd	Nd	Nd	Nd	Nd	Nd
N4	(10-12 cm)	3.23x10 ⁻¹⁵	1.82x10 ⁻¹⁵	2.52x10 ⁻¹⁵	1.48x10 ⁻²	8.35x10 ⁻³	1.16x10 ⁻²
	(12-14 cm)	Nd	Nd	Nd	Nd	Nd	Nd
	(14-16 cm)	Nd	Nd	Nd	Nd	Nd	Nd
	(16-18 cm)	Nd	Nd	Nd	Nd	Nd	Nd
	(18-20 cm)	Nd	Nd	Nd	Nd	Nd	Nd
N5	(10-12 cm)	Nd	Nd	Nd	Nd	Nd	Nd
	(12-14 cm)	Nd	Nd	Nd	Nd	Nd	Nd
	(14-16 cm)	7.05x10 ⁻¹⁶	3.97x10 ⁻¹⁶	5.49x10 ⁻¹⁶	3.24x10 ⁻³	1.82x10 ⁻³	2.52x10 ⁻³
	(16-18 cm)	Nd	Nd	Nd	Nd	Nd	Nd
	(18-20 cm)	Nd	Nd	Nd	Nd	Nd	Nd

Table 8. Calculated I-131 values by using the ratios given in Table 7

CONCLUSION

As a result, at the end of the experimental studies, literature data and calculations, I-131 activity in the Bafa Lake sediments was found between 9.78 x10⁻³ and 1.02x10⁻³ Bgkg⁻¹. The average I-131 activity in the Bafa Lake sediments was found to be 2.1×10^{-3} Bqkg⁻¹.

According to the measurements made in May 1986, the region with the highest concentration of I-131 radioactivity in Turkey was Edirne and its vicinity. In the first month after the accident, due to the limited data on I-131 radioactivity measurements in the Eastern Black Sea Region, a region-specific iodine assessment could not be made. Studies are being made to solve the deficiency of I-131 radioactivity measurements that are valid for many countries during the Chernobyl accident.

In this study, it was found that I-131 radioactivity can be predicted using the amount of I-129 radioisotope, a very long half-life fission product.

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Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Author's Contributions

The authors declare that they have contributed equally to the article.

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